Magnetic structure of copper-manganese alloys with exchange anisotropy

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We investigate the magnetic hysteresis and the residual magnetization of the alloy 77.6 at. % Cu + 22.4 at. % Mn in a wide temperature range from 4.2 to 250°K in magnetic fields up to 50 kOe. The dependence of the shift of the hysteresis loop observed in the given alloy on the residual magnetization is obtained. It is shown that at temperatures $T < T_c$ there exist in the alloy two ferromagnetic phases and possibly an antiferromagnetic phase. The Curie temperatures of these phases are estimated. It is established that the shift of the hysteresis loop is linearly connected with the reciprocal of the residual magnetization, thus confirming the hypothesis that ferromagnetic clusters are present in the alloy. The magnetic moments and the volumes of these clusters are estimates from the shift of the hysteresis loop and from the temperature at which an abrupt decrease takes place in the residual magnetization.

Meiklejohn and Bean [1,2] first observed in the systems Co and CoO the phenomenon of exchange anisotropy, the existence of which they attributed to exchange interaction on the boundaries of the ferromagnetic and antiferromagnetic regions. Exchange anisotropy gives rise to an asymmetrical hysteresis loop. It was shown later [3–11] that an asymmetrical hysteresis loop is observed also for many other substances, including solid solutions of Mn with Cu and Ag.

From our investigation of the Cu–Mn alloys containing from 5.7 to 22.4 at.% manganese [10,11] it follows that the largest residual magnetization of the shifted hysteresis loop is observed in the alloy with 22.4 at.% Mn and 77.6% Cu. We have therefore undertaken a detailed study of exchange anisotropy in just this alloy.

EXPERIMENTAL RESULTS

We investigated the temperature dependences of the magnetization, of the hysteresis loop, and of the residual magnetization in magnetic fields up to 50 kOe at temperatures 4.2–250°K, and also the dependence of the shift of the hysteresis loop on the residual magnetization. The magnetization was measured by a ballistic method using a type F-18 microweber meter. The difference between the number of turns of the differential measurement coils was 3300. The sensitivity of the apparatus was 0.1 G/div.

Figures 1 and 2 show the magnetization curves and the hysteresis loops for a quenched alloy at temperatures from 12.2 to 133°K. It is seen from the figures that with increasing temperature the residual magnetization and the shift of the hysteresis loop decrease. At 30°K and above, the asymmetry vanishes and the hysteresis loops have the shape usually possessed by ferromagnets. In stronger fields and at 60–70°K and above, the magnetization phenomena in the investigated fields are reversible. Typical of the region of relatively strong fields are broad gently-sloping sections of a stretched-out hysteresis loop.

Figure 3 shows the temperature dependence of the residual magnetization I_r after application of a field $H_{max} = 46$ kOe and cooling the sample to 4.2°K in different magnetic fields H_{cool}.

Figure 4 shows the hysteresis loops of a sample cooled in a magnetic field $H_{cool} = 1$–5 kOe, 2–25 kOe, and 3–46 kOe. It is seen from the figure that the residual magnetization increases with increasing magnetic field, and the hysteresis loops in these magnetic fields become narrower and its shift decreases.

DISCUSSION OF RESULTS

It is presently regarded as established (see [3–4] and [11]) that in Cu-Mn alloys, as well as in others, concen-

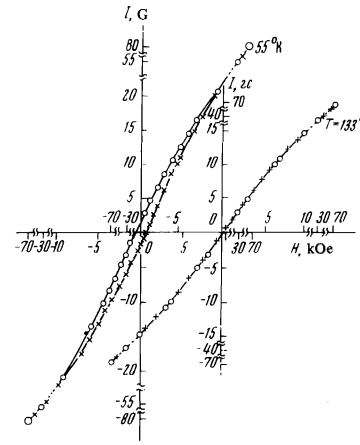

FIG. 1. Magnetization curves at 55 and 133°K.

tration inhomogeneities in small volumes make ferro- and antiferromagnetic ordering possible. Thus, in small regions at certain average distances between the Mn ions, the indirect exchange interaction via the conduction electrons leads to ferromagnetic ordering. Such regions are called "clusters." On the boundary between the cluster and its surrounding antiferromagnetic region there exists a strong exchange interaction, which orients the spins of the cluster along the local direction of the antiferromagnetism vector.

Cooling of the alloys in strong magnetic fields (above 40 kOe) orients the antiferromagnetic and ferromagnetic regions along the field, as a result of which a strong unidirectional anisotropy appears and leads to a shift of the hysteresis loop relative to the origin in the direction of the applied field. Cooling in a strong magnetic field can increase the residual magnetization at $T = 4.2$°K, as seen from Fig. 4, by 2–2.5 times, and this confirms the hypothesis that the magnetic moments of the clusters are oriented along the field.

At a certain temperature T_c, the orientation of the magnetic moments of the system of clusters is destroyed, and the system goes over into the supermagnetic state. At low temperatures, the clusters make the principal contribution to the residual magnetization I_r of the alloys, so that T_c can be experimentally determined from the temperature dependence of $I_r(T)$.

It is seen from Fig. 3 that the decrease of the residual magnetization I_r in the course of heating of a sample previously cooled in a magnetic field proceeds in two stages. In the temperature interval 10–20°, a rapid decrease of I_r is observed and can be attributed to the transition of the cluster system into the superparamagnetic state. The perpendicular sections on the hysteresis curve will then disappear, but the residual

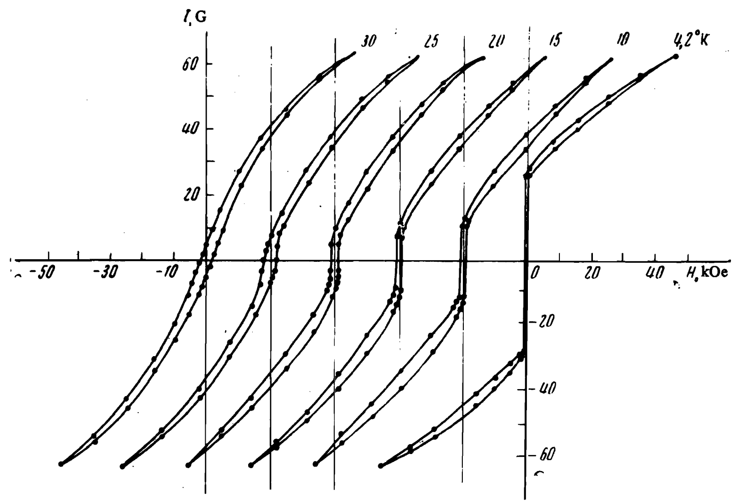

FIG. 2. Hysteresis loops obtained at temperatures 30, 25, 20, 15, 10, and 4.2°K after cooling the alloy to 4.2°K in a magnetic field of intensity 46 kOe.

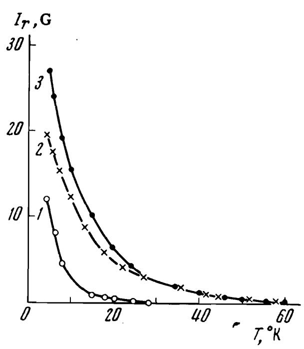

FIG. 3. Temperature dependence of residual magnetization of alloy after cooling to 4.2°K in the magnetic field: 1—H_{cool} = 0, 2—25 kOe, 3—46 kOe.

magnetization does not vanish completely. The rapid decrease of the residual demagnetization is followed by a slow decrease of the stable part of I_r in the interval from 20–30° to approximately 60°K.

At temperatures higher than 30°K, the shift of the hysteresis loop and the perpendicular sections are not observed, the broad gently-sloping sections come closer together, and the hysteresis loop assumes the form usual for a "normal" ferromagnet (see Fig. 1). The existence of broad gently-sloping sections of the hysteresis curve in the region of relatively strong fields and of a residual magnetization that is stable at temperatures above 30°K indicates that the alloy contains in addition to the cluster (ferromagnetic) and antiferromagnetic phases also a normal ferromagnetic phase.

Extrapolating the steep part of the $I_r(t)$ curves to the temperature axis, we can determine the temperature T_c at which the residual magnetization of the cluster ferromagnetic phase vanishes (the cluster become superparamagnetic), and from the temperature at which the "stable" residual magnetization vanishes we can determine the Curie temperature of the second ferromagnetic phase. The temperatures T_c coincide with the temperatures at which the cluster ordering disappears from the hysteresis loops, while T_r coincides with the temperature at which the magnetization curve becomes reversible. We see that the values of the temperatures T_c and T_r increase if the sample is cooled in a magnetic field, but these temperatures become constant starting with a definite field H_{cool} and higher.

The product $\mu_c \Delta \overline{H}_c$, where ΔH_c is the shift of the hysteresis loop and μ_c is the magnetic moment of the cluster, characterizes the average interaction energy of the cluster spins with the antiferromagnetic phase. It is therefore reasonable to write the connection be-

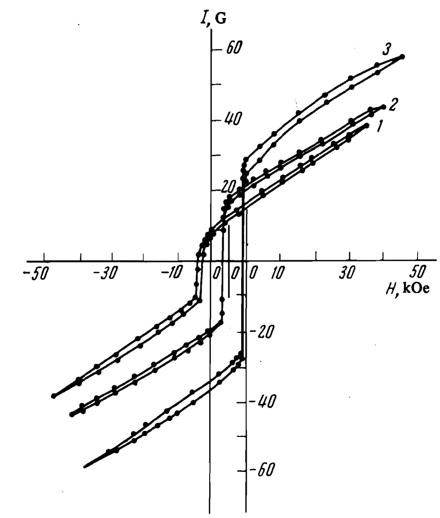

FIG. 4. Hysteresis loops obtained after cooling the sample to 4.2°K in magnetic fields of intensity 5 kOe (1), 25 kOe (2), and 46 kOe (3).

tween H_c and T_c in the form

$$\mu_c \Delta \overline{H}_c = kT_c, \quad (1)$$

where T_c is the temperature at which the residual magnetization of the cluster vanishes and k is the Boltzmann constant. Formula (1) makes it possible to determine, from the experimentally obtained values of ΔH_c and T_c, the values of the longitudinal components of μ_{cH} of the magnetic moment μ_c. The values of μ_{cH} obtained in this manner range from 100 to 700 μ_B, depending on the magnetic treatment of the alloy. These values are close to the values of μ_c obtained by Beck [12] for an alloy of 75% Cu + 25% Mn from the magnetization curves at high temperatures.

We have shown earlier that the values of I_r depend on the heat treatment of the alloy [11]. Figure 5 shows the dependence of the cluster magnetic-moment component μ_{cH} on the residual magnetization I_r. This dependence turned out to be linear, thus indicating a connection between I_r and the degree of orientation of the magnetic moments of the clusters. With increasing cooling field, the residual magnetization I_r increases, whereas the shift ΔH_c decreases accordingly (Fig. 6). The product of the residual magnetization by ΔH_c remains constant. Thus, the value of ΔH_c is also connected with the degree of orientation of the magnetic

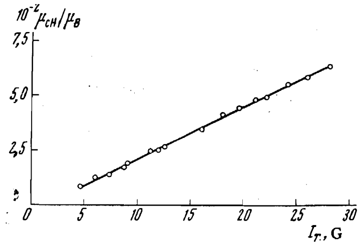

FIG. 5. Dependence of the magnetic moments μ_{cH} of the clusters on the residual magnetization I_r at 4.2°K, following different magnetic annealing procedures.

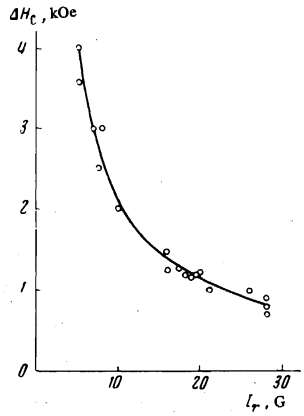

FIG. 6. Dependence of the displacement field on the residual magnetization at 4.2°K after different thermal magnetic treatments.

moments of the clusters. Indeed, if $\overline{\cos\alpha}$ is the mean value of $\cos\alpha$, where α is the angle between the direction of μ_c and H, then we have

$$I_r \sim \overline{\cos\alpha}, \quad \Delta H_c \sim 1/\overline{\cos\alpha}, \quad \Delta H_c I_r \approx \text{const}.$$

The magnetic moment is closest to the component $(\mu_{cH})_{max}$ corresponding to the largest value of I_r. Assuming $\mu_c \approx (\mu_{cH})_{max}$, we can estimate the average volume of the cluster with the aid of the formula

$$\mu_c = \mu_{Mn}\, n\, v_c, \tag{2}$$

where v_c is the volume of the cluster, n is the concentration of the manganese atoms, μ_{Mn} is the magnetic moment of the manganese atom. At a maximum residual magnetization the volume of the cluster calculated from formula (2) turns out to be 10^{-20} cm^3, and accordingly its transverse dimension l_c is of the order of 21×10^{-8} cm. The value of μ_{Mn} was assumed equal to the magnetic moment of the free Mn ion as calculated from the susceptibility in the paramagnetic region.

CONCLUSION

Thus, we have demonstrated the possibility of determining the magnetic moment and the volume of ferromagnetic clusters from the experimentally obtained values of the displacement of hysteresis loop ΔH_c and the temperature T_c obtained by extrapolating the steep part of the magnetization curve $I_r(T)$ to the T axis.

We have also shown that in addition to ferromagnetic clusters and the antiferromagnetic phase, there exists in the investigated alloy a normal ferromagnetic phase, which makes a contribution of several gauss to the residual magnetization and has an appreciable coercive force on the order of 3 kOe. Part of the hysteresis loop in strong fields (above 20 kOe) is due to the irreversible magnetization of this phase. This phase interacts little with the antiferromagnetic regions and makes no essential contribution to the exchange anisotropy. Therefore, up to the temperature T_c the hysteresis loop is symmetrical about the origin in strong magnetic fields, and above T_c the entire hysteresis loop becomes symmetrical.

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240