Ferromagnetic ordering in systems of magnetic moments of super-paramagnetic particles

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The ferromagnetic alignment of magnetic moments of dispersed (~ 30Å) ferromagnetic particles couched in a nonferromagnetic matrix was observed.

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Transitions from the super-paramagnetic (SPM) state to macroscopic glass are observed upon lowering the temperature in systems of dispersed, equiaxial, ferromagnetic metallic impurities (average-size ~ 100 Å, density $n \approx 10^{17}$ cm^{-3}) located in a nonferromagnetic metallic matrix.[1] In the case of alloys,[1] the mean distance between adjacent particle surfaces measured along a line through their centers, $\xi > d$.

Reducing the interparticle spacing ($\xi \leqslant d$) may change the nature of the transition since in this case a change in the type of coupling between the particles is possible.

As a test object we picked a system of solid-solution impurities consisting of a ferromagnetic compound Cu_2MnAl in the Cu_3Al matrix. The particles were formed as

a result of decomposition of the supersaturated solid solution containing Cu–62.5, Mn–12.5 and Al–25 (%at.) (composition corresponding to a fraction $p = 0.3$–0.4, of the ferromagnetic phase by volume[2]). After homogenation, specimen were heated to 1123 K in water and annealed at 523 K for 20 sec and 4 min.

Monitoring of the structural states—carried out by means of the x-ray diffusion scattering method—identified the presence in the thermally-treated specimen of equiaxial particle systems, the dilation centers. The parameters d, ξ, and n of these specimen are tabulated below

TABLE I.

Burn-off time	d, Å	ξ, Å	M, μ_B	T_1, K	T_2, K	n, cm^{-3}
20 sec –1	28	15	1.1×10^3 ($T = 273$ K)	223	340	1.2×10^{19}
4 min –2	42	25	4.5×10^3 ($T = 393$ K)	—	600	0.45×10^{19}

In the selection of the above alloy we were guided by a need to satisfy the condition $KV \ll kT$ at $T \geqslant 77$ K, where K is the magnetic anisotropy constant, V is the mean particle volume and k is Boltzmann's constant. In the case of Cu_2MnAl $K = 10^3$ erg/cm^3,[3] and this condition is also satisfied for particles with sizes greater than those used in our work.

Figures 1 and 2 show the temperature dependence of the magnetic properties of the aforementioned particle systems (states with a smaller d are designated 1, with greater 2). Clearly, the two structural states correspond to a different dependence of the low-field magnetic susceptibility $\chi(T)$. ($\chi(T)$ was measured by means of an induction method at the frequency and amplitude of the measuring field, 50 Hz, and ~ 5 Oe, respectively). If $\chi(T)$ for the state 2 follows a curve with a maximum (Fig. 2 curve 1) which is characteristic for a state of the type of spin glass,[1] the dependence in Fig. 1 is typical for a bulky ferromagnetic: an insignificant increase in the susceptibility as the temperature increases from 77 K followed by a sharp decrease at certain temperature T_1.

Figure 1 also shows the temperature dependence of specimen magnetization I, measured in a 9-kOe field–$I_s(T)$ (the solid line connects experimental points and its dotted extension was obtained by extrapolation). The experimental value of I_s follow a straight line constructed in the I_s^2-T coordinates. The straight line intersects the abscissa at temperatures $T_2 > T_1$ ($T_2 - T_1 = 120°$), and it may be regarded as the Curie temperature of material particles T_K. (Measurements for this specimen were carried out at temperatures not exceeding 300 K in order to avoid decomposition of the solid solution during measurement).

Curves $I(T)$ with maxima,[1] correspond to state 2 (macroscopic glass type) in small fields and state 1 is characterized by functions $I(T)$ that are typical for ferromagnetics. The dependence of H/σ on σ^2 was shown graphically (Fig. 3, $\sigma = I/\rho$, ρ is the material density). At temperatures near T_1 the experimental data points lie along

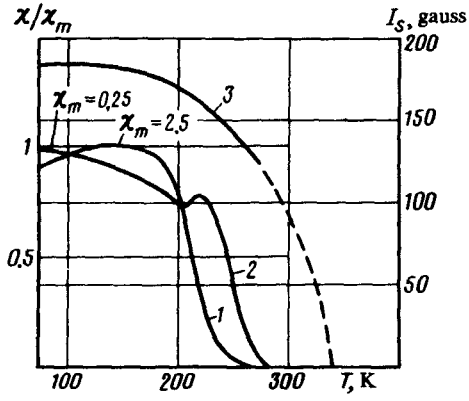

FIG. 1. Temperature dependence of magnetization and magnetic susceptibility of specimen in state 1: curve 1—low-field susceptibility χ/χ_m (where χ_m is maximum value in electromagnetic units); curve 2—susceptibility measured in a constant field $H = 100$ Oe; curve 3—magnetization in a field $H = 9$ kOe.

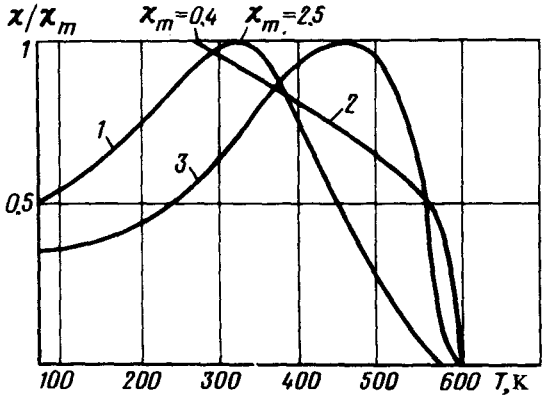

FIG. 2. Temperature dependence of magnetic susceptibility of specimen in state 2: curve 1—low-field magnetic susceptibility (burn-off time $\tau = 4$ min); curve 2—susceptibility χ in a constant field $H = 100$ Oe ($\tau = 4$ min); curve 3—low-field susceptibility ($\tau = 16$ min).

straight lines, with the line corresponding to T_1 crossing the origin of the coordinate system (Fig. 3). Consequently, a phase transition of the second kind occurs at T_1 which should be characterized by a linear dependence of H/σ on σ^2.[4] Assuming that the relationship $H/\sigma = \alpha + \beta\sigma^2$ (α and β are constants) describes the process of natural magnetization (paraprocess) and that, according to Fig. 3, the linear dependence of H/σ on σ^2 occurs in fields $\geqslant 70$ Oe, it may be concluded that the differential magnetic susceptibility measured in a field $H = 100$ Oe shall represent the susceptibility of the paraprocess χ_p. The dependence of χ_p on T for state 1 is shown in Fig. 1 (curve 2). (A constant field was applied in parallel to the measured, the coersive force $H_c \leqslant 1$ Oe at $T \to T_1$).

The presence of a maximum in the paraprocess susceptibility serves as an additional confirmation of the fact that a phase transition occurs at $T = T_1$. In the temperature range $T_1 < T < T_2$ a specimen is in the SPM state and at $T = 273$ K the particle magnetic moment $M \approx 10^3 \mu_B$, where μ_B is the Bohr magneton (see Table I).

According to the experimental data above, the following sequence of magnetic states corresponds to a specimen in state 1: at $T > T_2$—paramagnetic, $T_1 < T < T_2$—

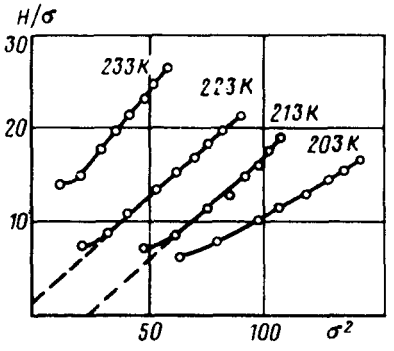

FIG. 3. Magnetization isotherms of specimen in state 1.

superparamagnetic, and at $T < T_1$—ferromagnetic. It should be assumed that at $T \leqslant T_1$ ferromagnetic ordering in a system of magnetic moments of SPM particles takes place.

Increasing the burn-off time (state 2) leads to a rise in the Curie temperature ($T \to T_K$ Cu_2MnAl) as evidenced by the temperature dependence of χ_p for this state measured in a 100-Oe field. A sharp decrease in χ_p occurs as $T \to T_K$ due to a decrease in the particle magnetic moment. A rise in the T_K may be associated with changes in the concentration of Mn atoms and a possible increase in the long-range atomic order in particles. Subsequent increases in the burn-off time to 16 min fail to noticeably change T_K but result in increased temperature of transition into the macroscopic glass state in accordance with Ref. 1.

It may be assumed that the change in the type of magnetic state as d and ξ increase depends on the fact that, for state 1, a coupling due to conductivity electrons is significant. There are reasons that admit its existence. Spin polarization of the conductivity electrons which occurs in a ferromagnetic extends[5] to adjacent nonferromagnetic metal regions. The degree of polarization decreases exponentially with distance from the ferromagnetic material surface, the effective penetration depth depending on the free path of the conductivity electrons.[5]

At small ξ (the electron cloud polarized in a single particle reaches the adjacent impurities) an energy-specific situation clearly develops when the direction of the magnetic moment of an impurity coincides with the direction of spin of the conductivity electrons polarized in the adjacent particle. The system of interacting impurities sustains therefore the aforementioned phase transition which at $T < T_1$ results in the ferromagnetic alignment of their magnetic moments.

A small increase in ξ may lead to the predominance of a contribution from the dipole-dipole coupling that causes the formation of a magnetic state of the macroscopic glass type.

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[1]V.V. Kokorin and A.E. Perekos, Pis'ma Zh. Eksp. Teor. Fiz. **27**, 500 (1978) [JETP Lett. **27**, 470 (1978)].

[2]M. Bouchard and G. Thomas, Acta met. **23** 1485 (1975).
[3]K. Aoyagi and M. Sugihara, J. Phys. Soc. Japan **16**, 1027 (1961).
[4]K.P. Belov, Magnitnye prevrashcheniya (Magnetic Conversions), M., 1959.
[5]V.Ya. Gamlitskiĭ, O.A. Gurkovskiĭ, V.I. Nikolaev, I.N. Nikolaev, V.M. Cherepanov and S.S. Yakimov, Zh. Eksp. Teor. Fiz. **67**, 756 (1974) [Sov. Phys. JETP **40**, 374 (1975)].