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Long-range Ferromagnetic Order in Ensemble of Nanometric Magnetic Particles with Giant Magnetic Moments

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Abstract

Using X-ray and magnetic methods as well as small-angle neutron scattering, it is shown as an example of the decomposing alloy $Cu_{64}Mn_9Al_{27}$ that the appearance of long-range ferromagnetic order in a system of small superparamagnetic particles dissolved in nonmagnetic metallic matrix is due to cooperative ordering of their magnetic moments.

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1 Introduction

In the model of localized magnetic moments the appearance of magnetically ordered, specifically ferromagnetic (FM) state, in metals and alloys of $3d$-transition metals is usually attributed to the spin ordering of uncompensated atomic magnetic moments whose magnitudes do not exceed several Bohr magnetons μ_B [1]. It is interesting to determine whether or not long-range FM order (LRFMO) can appear in system of magnetic moments ranging from several hundreds to several thousands μ_B.

In the present work this question is solved as an example of the decomposing alloy $Cu_{64}Mn_9Al_{27}$ on the basis of investigations of the critical neutron scattering in combination with structural and magnetic investigations.

2. Experimental

The alloy $Cu_{64}Mn_9Al_{27}$ was smelted in an induction furnace in argon atmosphere from chemically pure copper, manganese and aluminium.

A mutual-induction bridge was used to measure the real χ' and imaginary χ'' components of AC susceptibility. DC magnetization was investigated with a standard vibrating-sample magnetometer. X-ray study was performed by photo method in an X-ray chamber with oscillation of a sample. All these experiments were carried out in the Institute for Magnetism of National Academy of Sciences of Ukraine, Kiev. The small-angle neutron scattering (SANS) experiments were performed on PAXE spectrometer in Léon Brillouin Laboratory, Saclay, France.

3. Results and discussion

3.1. Magnetic and structural characteristics of $Cu_{64}Mn_9Al_{27}$ alloy

First it is necessary to examine the magnetic and structural characteristics of the alloy $Cu_{64}Mn_9Al_{27}$ after different heat treatments. According to X-ray investigations the quenched alloy $Cu_{64}Mn_9Al_{27}$ is a homogeneous solid solution with Cu_3Al-type structure (unit cell lattice parameter a = 2.986 Å). The Mn atoms are randomly dissolved in the matrix of the alloy. Therefore it can be expected that at low temperatures, on account of the presence of indirect Ruderman–Kittel– Kasuya–Yoshida (RKKY) exchange, a spin-glass (SG) state will arise in the alloy, as happens in the classical SG system CuMn [2]. It is obvious from Fig. 1a, which displays the temperature dependence of real component χ' of the dynamic magnetic susceptibility of the alloy in quenched to the ice water from temperature 1050 K state, that at the temperature $T_G = 40.2$ K the alloy indeed passes from a paramagnetic (PM) into a SG state. However, the alloy decomposes as a result of annealing at temperature $T_{AN} = 373$ K for 5 h. Its structural and magnetic characteristics change substantially. In what follows, we shall be concerned with this state.

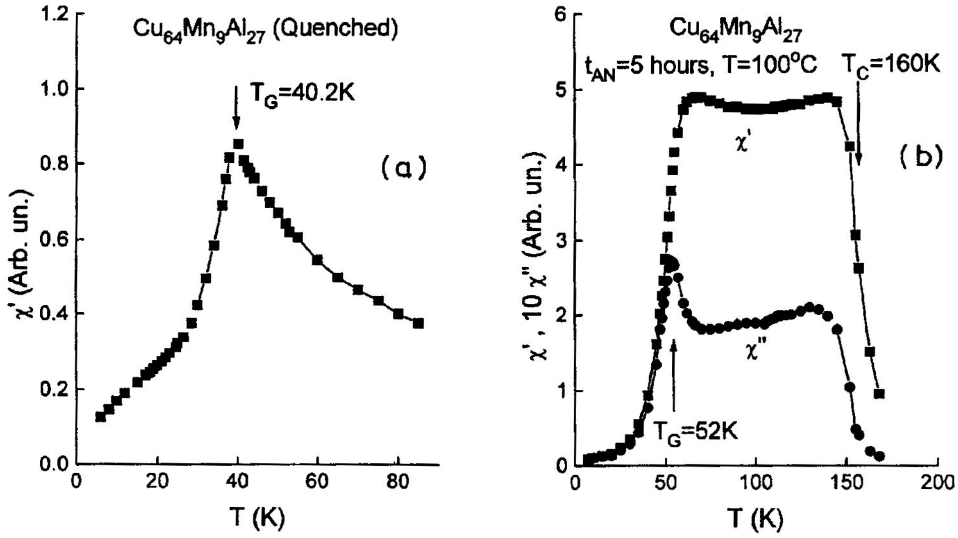

Fig. 1. Temperature dependences of the real χ' and imaginary χ'' components of AC susceptibility of the alloy $Cu_{64}Mn_9Al_{27}$ in quenched (a) and decomposed (b) state.

The investigations of diffuse scattering of X-rays near the Bragg reflections of the matrix show that there are one pair of satellites near the (200) reflection and two pairs near (110) one (see Fig. 2). Such a type of satellites is typical of isomorphic decomposed solid solutions that contain an ensemble of coherent equiaxed particles of precipitated phase. Being the centers of dilatation, these particles are homogeneously dissolved in anisotropic elastic matrix. The number of satellites pairs vs. Miller indexes of Bragg reflections points out that particles are situated in the matrix more or less regularly along ⟨100⟩ direction [3].

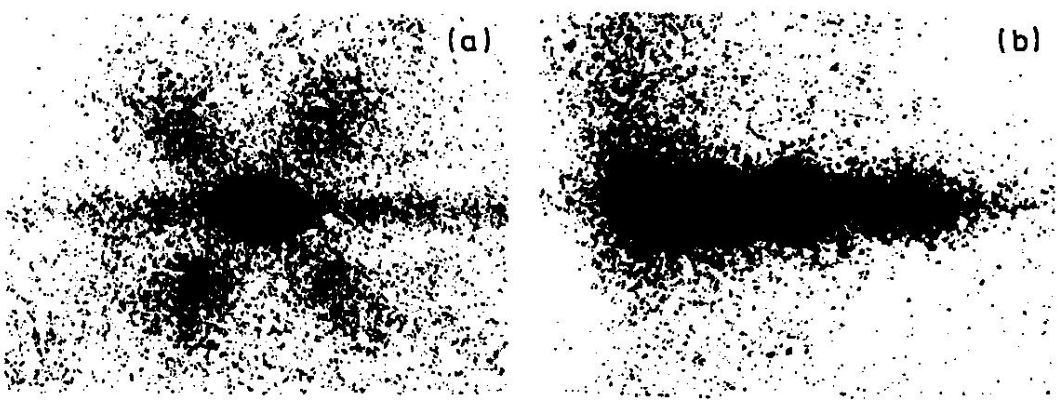

Fig. 2. The regions of X-ray diffusion scattering for the decomposed alloy $Cu_{64}Mn_9Al_{27}$ near (110) (a) and (200) (b) Bragg reflections.

Using the picture of diffuse scattering it is possible to estimate the mean distance D between centers of particles. According to [4] the distance D corresponds to periodical variation of lattice parameter of matrix and may be estimated from the formula

$$D = a\frac{h(\tan\theta)}{(h^2+k^2+l^2)\delta\theta_1},$$

where $h,\ k,\ l$ are the Miller indexes, and a is the lattice parameter of matrix, θ is the Bragg angle of (hkl) reflection, $\delta\theta_1$ is the angular distance between centers of satellites. In addition to that it is possible to estimate the mean size d of the particles. According to [3]

$$d = \frac{\lambda}{2\delta\theta_2\cos\theta},$$

where λ is the wavelength of X-rays and $\delta\theta_2$ is the angular width of a satellite pair in reciprocal space. The results of calculations of D and d are presented in Table. Assuming that in the decomposition process particles with the stoichiometric composition Cu_2MnAl (Heusler's alloy) are formed and knowing the lattice parameter $a = 5.971$ Å, the number of Mn atoms per unit cell $N = 4$, and the magnetic moment of the Mn atom $\mu = 4\mu_B$ [5] in the newly formed phase, it is easy to find the magnetic moment of the separated particles (see Table).

Table Structural and magnetic characteristics of decomposed alloy $Cu_{64}Mn_9Al_{27}$.

Parameters	X-ray data	Magnetic data
d[Å]	30 ± 3	32 ± 2
D [Å]	47 ± 3	51
μ [μ_B]	1100 ± 320	1140 ± 150
N [10^{18} cm^{-3}]	8.84	7.34
Volume fraction	0.136	0.126

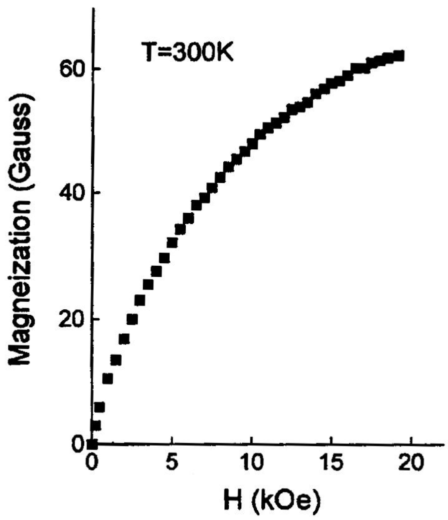

Fig. 3. DC magnetization of the decomposed alloy $Cu_{64}Mn_9Al_{27}$ at temperature 300 K.

Investigations of the static magnetization at room temperature in magnetic fields up to 20 kOe show that the alloy is a typical superparamagnet (SPM) (see Fig. 3). Indeed, anisotropy constant is $K = 10^3$ erg/cm^3 for bulk alloy Cu_2MnAl at room temperature [5]. Therefore, the magnetic energy of particle with $d = 30$ Å is $KV/k_B = 0.26$ $K \ll 300$ K and the condition $KV \ll k_BT$ of SPM behavior fulfills very well. Here V is the volume of the particle. Using the known relation of the theory of SPM [1]

$$M = \frac{N\mu^2 H}{3k_BT} \quad \left(\frac{\mu H}{k_BT} \ll 1\right), \qquad M = N\mu\left(1 - \frac{k_BT}{\mu H}\right) \quad \left(\frac{\mu H}{k_BT} \gg 1\right),$$

and experimental data from Fig. 3 and assuming that at room temperature the spontaneous magnetization of the particles is equal to the value $M_S = 500$ Gs for the bulk alloy Cu_2MnAl [5], it is easy to estimate their average magnetic moment μ, size d, concentration N, and other parameters. Comparing the results obtained by X-ray and magnetic methods shows that they are in good agreement with each other (see Table).

The temperature dependences of the real χ' and imaginary χ'' components of the dynamic magnetic susceptibility of the decomposed alloy $Cu_{64}Mn_9Al_{27}$ are displayed in Fig. 1b. It is evident from the data presented that the alloy undergoes a double temperature transition PM–FM–SG. In the present work, however, we shall not examine in any detail the reentrant transition FM–SG at $T_G = 52$ K. As mentioned above, our main objective is to determine the nature of the appearance of LRFMO at the Curie temperature $T_C = 160$ K.

3.2. Critical neutron scattering

Since FM ordering arises in the experimental alloy only in the decomposed state, it is natural to conjecture that the elementary carriers of magnetism in this case are not the separate magnetic moments of Mn atoms but rather SPM particles at temperatures $T > T_C$ with effective magnetic moment $\mu \approx 10^3\mu_B$. Evidently, in the latter case, under the conditions of FM ordering of the magnetic moments of an ensemble of such particles, the radius R_C of the FM critical fluctuations near T_C should satisfy the condition $R_C \geq \xi \approx D - d$, where ξ is the distance between the

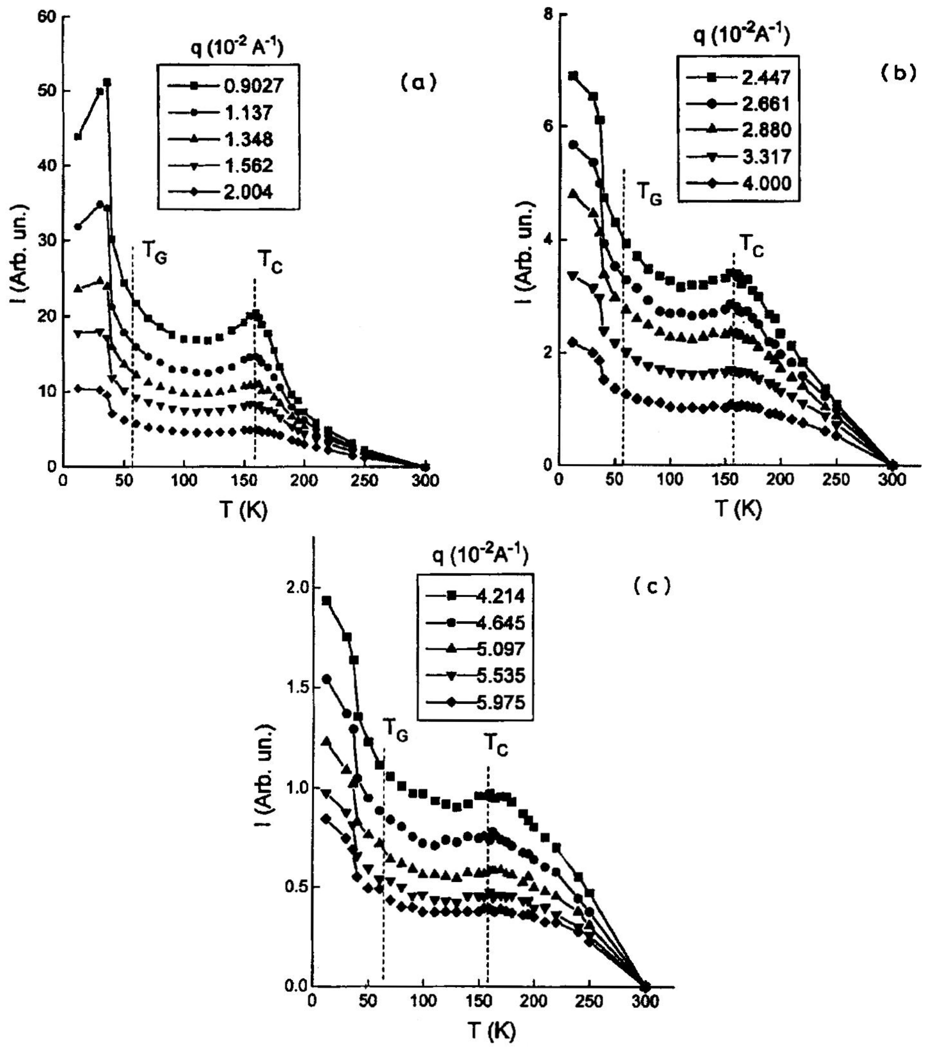

Fig. 4. Temperature dependences of the intensity I of small-angle neutron scattering for the decomposed alloy $Cu_{64}Mn_9Al_{27}$, for various $\boldsymbol{q}$ vectors ($\boldsymbol{q}$ is the wave vector of the scattering neutrons).

surfaces of the particles along the line connecting their centers. In other words, critical scattering peaks should be observed in the temperature dependences of SANS in the temperature range $TC = 160\ K < T_C^*$, where $T_C^* \approx 700$ K is the local Curie temperature of the SPM particles, only for neutron wave vectors $q \leq R_C^{-1} = \xi^{-1} \approx 0.05$ Å^{-1}.

The experimental data presented in Fig. 4 fully confirm the arguments presented above. Indeed, a critical neutron scattering peak is clearly recorded at temperature $T_C = 160$ K for neutron wave vectors $q \leq 0.05$ Å^{-1}. At the same time, there are no appreciable anomalies in the temperature dependences of the intensity of the scattered neutrons for $q > 0.05$ Å^{-1}.

The following should be noted concerning the interactions which result in ordering of mentioned above type. Estimates show that for the values found for the parameters of the system of precipitated particles (see Table), the dipole interparticle interaction gives a Curie temperature $T_C \approx 6-100$ K, which is lower than the experimentally determined value. Apparently, in the case considered, on account of the quasi-regular arrangement of the precipitates in the matrix of the alloy, RKKY superexchange between particles plays the main role in the formation of the FM, just as in multilayer structures of the type Co/Cu/Co or Fe/Ag/Fe [6]. In the latter case, the alternating magnetic and nonmagnetic layers have approximately the same thickness as those found in the present work (d and ξ).

In addition, it must be noted that as a result of decomposition of the alloy investigated above, only 35% of the Mn atoms participate in the formation of the stoichiometric FM phase Cu_2MnAl. The rest of the Mn atoms remains dissolved in the nonmagnetic matrix. As was stressed in [7], such "lost spins" can also play an important role in the formation of LRFMO in heterogeneous magnetic systems.

4. Conclusion

It has been shown in the present work on the example of the alloy $Cu_{64}Mn_9Al_{27}$ that long-range ferromagnetic order can arise in system with small SPM particles, which appear as a result of isomorphic decomposition of the alloy.

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