ISSN 1063-7834, Physics of the Solid State, 2007, Vol. 49, No. 9, pp. 1740–1743. © Pleiades Publishing, Ltd., 2007.
Original Russian Text © S.G. Barsov, S.I. Vorob'ev, V.P. Koptev, S.A. Kotov, S.M. Mikirtych'yants, G.V. Shcherbakov, 2007, published in Fizika Tverdogo Tela, 2007, Vol. 49, No. 9, pp. 1660–1663.

MAGNETISM AND FERROELECTRICITY

Investigation of the Magnetic Properties of Homogeneous Copper–Manganese Alloys

S. G. Barsov, S. I. Vorob'ev, V. P. Koptev, S. A. Kotov, S. M. Mikirtych'yants, and G. V. Shcherbakov

Konstantinov Institute of Nuclear Physics, Russian Academy of Sciences, Gatchina, Leningrad obtast, 188300 Russia
e-mail: vsiloa@pnpi.spbu.ru

Received November 8, 2006; in final form, February 22, 2007

Abstract—The magnetic characteristics of homogeneous copper–manganese alloys $Cu_{1-x}Mn_x$ are studied by the muon spin relaxation technique for the first time. It is revealed that the specific magnetic phase, which is most likely characterized by a fast spin dynamics and the absence of long-range order, is formed in alloys with concentrations $0.2 < x < 0.7$ in the temperature range 10–330 K. The complete magnetic phase diagram is constructed.

PACS numbers: 76.75.+i, 75.20.Hr, 75.50.Lk

DOI: 10.1134/S1063783407090211

1. INTRODUCTION

The $Cu_{1-x}Mn_x$ binary alloy belongs to materials that are characterized by a clear correlation of collective processes in crystalline and magnetic subsystems [1]. In these materials, there exists a close relationship between the martensitic transformation of the crystal lattice and the antiferromagnetic ordering of magnetic moments of manganese atoms. For example, the temperatures of the martensitic and antiferromagnetic transitions in copper–manganese alloys virtually coincide with each other, even though they depend substantially on the manganese concentration and the degree of homogeneity of the alloy [2].

Numerous experimental findings generalized in the review by Vintaĭkin et al. [1] give grounds to believe that, in these materials, martensitic transformations (structural transitions of the diffusionless collective type) are induced by the anisotropy of the exchange interaction at temperatures $T < T_N$ (where T_N is the temperature at which there arises an antiferromagnetic ordering of Mn atoms). The particular interest expressed by researchers in the study of $Cu_{1-x}Mn_x$ binary alloys is explained by the fact that they are typical representatives of shape memory materials [3].

Banerjee and Majumdar [4] thoroughly investigated the magnetic properties of the $Cu_{1-x}Mn_x$ alloys by macroscopic methods over a wide range of Mn concentrations ($x = 0.05$–0.83). The experimental data obtained were used to construct the magnetic phase diagram of the $Cu_{1-x}Mn_x$ alloys. It was assumed that a superparamagnetic phase can exist in these materials.

In our earlier works [5, 6], the muon spin relaxation technique was used with advantage to examine disordered magnetic states and spin dynamic processes of non-spin-wave nature. The spin-glass state in the $Cu_{1-x}Mn_x$ alloys at concentrations $x < 0.1$ was investigated using the muon spin relaxation technique by Uemura et al. [7]. The alloys with higher concentrations of Mn magnetic atoms ($x > 0.1$) were studied by this technique in our previous work [8]. For the first time, it was assumed that there can exists a new magnetic phase transition before the transition to the spin-glass state. In the present work, we investigated the $Cu_{1-x}Mn_x$ alloys over a wide range of concentrations of Mn magnetic atoms. In this case, special attention was focused on the measurements of the concentrations themselves.

2. SAMPLE PREPARATION, EXPERIMENTAL TECHNIQUE, AND RESULTS

In this work, the magnetic properties of homogeneous copper–manganese alloys $Cu_{1-x}Mn_x$ were studied by the muon spin relaxation technique on the synchrocyclotron at the Konstantinov Institute of Nuclear Physics of the Russian Academy of Sciences (Gatchina, St. Petersburg, Russia) and at the Paul Scherrer Institute (Switzerland). Samples were homogenized by quenching in water after their heat treatment in a muffle furnace at a temperature of 1100 K for 100 h.

In our experiments, we measured the time distributions of positrons $N_e(t)$ that were formed as a result of the decay $\mu^+ \longrightarrow e^+ + \nu_e + \tilde{\nu}_\mu$ (the muon lifetime is $\tau_\mu \approx 2.2$ μs) and emitted in the direction of the initial muon polarization (polarized muon beams were used) in a time window $\Delta t \approx 4.5\tau_\mu$ after each muon was stopped in the sample, as well as the integrated yields of these positrons [9]. The time distributions were approximated by the function

$$N_e(t) = N_0[1 + a_0 G(t)]\exp(-t/\tau_\mu), \qquad (1)$$

where the normalization constant N_0 and the maximum asymmetry a_0 characterize the experimental conditions specific for each sample and do not depend on the muon depolarization. The muon spin relaxation function $G(t)$ determined from the time distribution N_e reflects the effect of local magnetic fields on the muon spin at the site of its stopping. In particular, we have $G(t) = 1$ in the absence of depolarization and $G(t) = 0$ for nonpolarized muons.

Figure 1 presents the normalized integrated yields of positrons for samples with different concentrations of manganese atoms $N_e(\text{norm}) = (n_e/n_0 - 1)/a_0$. This integrated yield does not depend on the sample geometry, parameters of the muon spin relaxation setup, and muon beam polarization and provides a general model-independent information on muon depolarization under local magnetic fields. The parameters n_0 and a_0 were determined at a temperature considerably higher than the temperature of the transition to the magnetically ordered phase.

Specifically, the normalized integrated yield $N_e(\text{norm})$ measured for the $Cu_{0.2}Mn_{0.8}$ sample at temperatures $T > 330$ K in zero magnetic field tends to unity. This circumstance suggests the absence of muon depolarization in the far paramagnetic range, in which the frequency of oscillations of electronic moments is too high (~10^{12} Hz) for their magnetic field to change substantially the muon polarization. The paramagnetic state is also indicated by the complete depolarization of muons in a relatively weak transverse external magnetic field of ~580 Oe. In the temperature range 320–290 K, the normalized integrated yield $N_e(\text{norm})$ changes drastically and then reaches a value of ~1/3. This suggests that the sample transforms into a magnetically ordered state with an isotropic (on a local, cluster, or domain level) orientation of static internal local magnetic fields. This behavior is in good agreement with the phase diagram previously proposed in [4], according to which the antiferromagnetic transition at $T_N \sim 300$ K occurs in a homogeneous alloy with the concentration $x = 0.8$. The normalized integrated yield $N_e(\text{norm})$ equal to 1/3 is retained to $T \approx 200$ K. With a further decrease in the temperature, the normalized integrated yield $N_e(\text{norm})$ decreases sharply almost to zero. This indicates that, in the given temperature range, there arises a strong dynamic depolarization of muons. The temperature dependence of the normalized integrated yield $N_e(\text{norm})$ in the range 200–20 K is characteristic of frustrated magnets, which undergo transition to a low-temperature spin-glass state through an intermediate magnetically ordered phase with a long-range order. In this case, the dynamic polarization is associated with the transformation of the magnetic structure in the transition range [5].

The temperature dependences of the normalized integrated yield $N_e(\text{norm})$ for the samples with the concentration $x = 0.17$, 0.25, and 0.45 are also plotted in Fig. 1. The normalized integrated yield $N_e(\text{norm})$ for these samples decreases drastically with a decrease in the temperature due to the strong depolarization of muons. Since the normalized integrated yield $N_e(\text{norm})$ decreases almost to zero, the inference can be made that the arising local magnetic fields have a fluctuation character and are rather high. Moreover, the normalized integrated yield $N_e(\text{norm})$ for the sample with the concentration of manganese atoms $x = 0.45$ initially decreases to a small value and then remains virtually unchanged over a wide temperature range (from 200 to 60 K). This suggests to some extent that, in the given temperature range, the sample is in a specific phase state characterized by a fast spin dynamics. As the temperature decreases ($T < 60$ K), the normalized integrated yield $N_e(\text{norm})$ increases gradually to ~1/3, which corresponds to the transition of the sample to the isotropic magnetic phase with a slow spin dynamics.

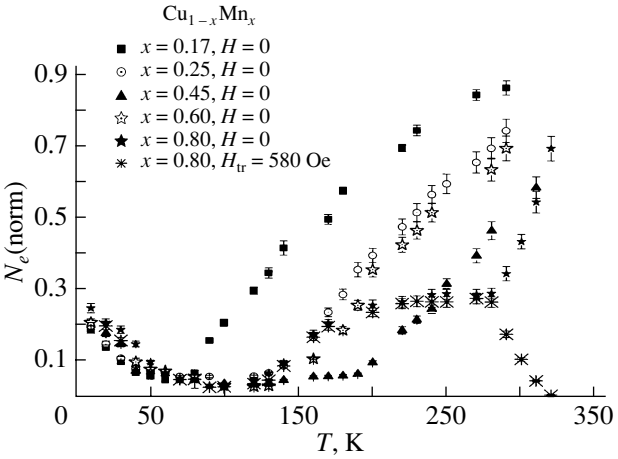

Fig. 1. Temperature dependences of the normalized integrated yield $N_e(\text{norm})$ of positrons for samples with different concentrations of magnetic atoms Mn_x.

It should be noted that the dependence of the normalized integrated yield $N_e(\text{norm})$ on the concentration x of magnetic manganese atoms exhibits one more feature. The rate of change in the normalized integrated yield $N_e(\text{norm})$ with a variation in the temperature in the high-temperature transition range is identical for all samples with concentrations $x = 0.17$–0.60. This indicates once again that the same physical processes occur in the alloys and that the characteristics obtained are not associated with the specific quality of the samples. The transition temperature increases with an increase in the concentration of magnetic atoms to $x = 0.45$. A further increase in the concentration of magnetic atoms is accompanied by a decrease in the transition temperature. For example, the transition temperature for the sample with the concentration $x = 0.60$ is approximately equal to that for the sample with the concentration $x = 0.25$. A similar dependence of the transition temperature on the concentration x was observed by Banerjee and Majumdar [4]. This effect was explained by the corresponding change in the size of antiferromagnetic clusters.

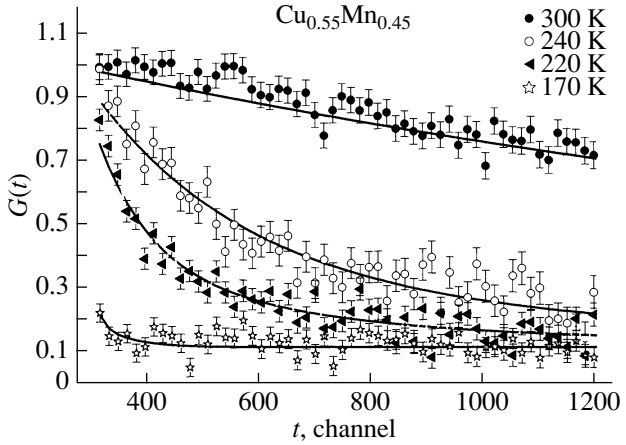

Fig. 2. Relaxation functions for the $Cu_{0.55}Mn_{0.45}$ sample at different temperatures in the range 170–300 K. One channel on the time scale corresponds to 0.625 ns. The origin of the scale is located at the 274th channel.

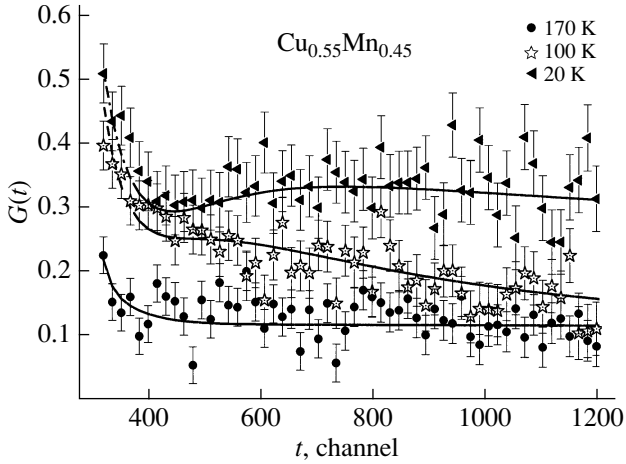

Fig. 3. Relaxation functions for the $Cu_{0.55}Mn_{0.45}$ sample at different temperatures in the range 20–170 K. One channel on the time scale corresponds to 0.625 ns. The origin of the scale is located at the 274th channel.

The analysis of the time spectra $N_e(t)$ demonstrates that the experimental data cannot be described using simple relaxation functions. In the paramagnetic range, the experimental data are well described by the relaxation function

$$G(t) = \exp(-\lambda t). \quad (2)$$

However, as the temperature of the first magnetic transition is approached (this can be judged from the drastic increase in the relaxation rate λ), the description of the experimental data requires the use of the relaxation function in the form of the sum of two exponential functions; that, is

$$G(t) = a_1\exp(-\lambda_D t) + a_2\exp(-\lambda t), \quad (3)$$

where $a_1 + a_2 = 1$, $G(t)$ is the dimensionless relaxation function varying from 0 to 1, and λ_D and λ are the dynamic relaxation rates for the corresponding exponential functions. The experimental data are best described using constant parameters $a_1 = 1/3$ and $a_2 = 2/3$.

At temperatures below 100 K, the experimental data for the samples can be described by the following relaxation function:

$$G(t) = [1/3 + 2/3(1 - \Delta t)\exp(-\Delta t)]\exp(-\lambda_D t). \quad (4)$$

For $\lambda_D \ll \lambda$, this form of the relaxation function G is consistent with the spin-glass model. In this case, the parameter λ_D corresponds to the relaxation associated with the occurrence of fluctuating random fields. The parameter Δ is connected to the static fields.

A more complex relationship for the relaxation function was proposed by Uemura et al. [7]. However, when describing the time spectra $N_e(t)$ for samples with high manganese concentrations ($x > 0.2$), the form of the relaxation function represented by expression (4) is more preferential.

The behavior of the relaxation functions for the $Cu_{0.55}Mn_{0.45}$ sample with a variation in the temperature is illustrated in Figs. 2 and 3. It can be seen from these figures that a virtually complete depolarization of the muon ensemble is observed in the temperature range 240–120 K. The relaxation function decreases only when the temperature of the sample decreases below a temperature of 100 K. At a temperature of 20 K, the relaxation function asymptotically approaches a value of ~1/3, which corresponds to the isotropic orientation of quasi-static local magnetic fields.

The temperature dependences of the parameters λ, λ_D, and Δ are shown in Fig. 4. There are two magnetic phase transitions. The first transition in the sample is observed at a temperature of ~200 K, and the second transition occurs at temperatures in the range 150–130 K.

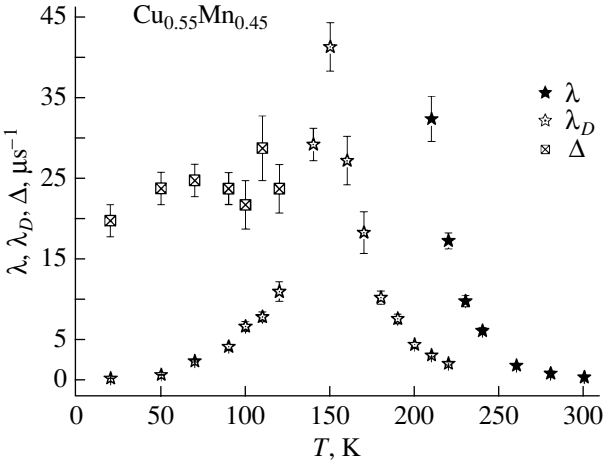

Fig. 4. Temperature dependences of the dynamic (λ, λ_D) and static (Δ) relaxation rates.

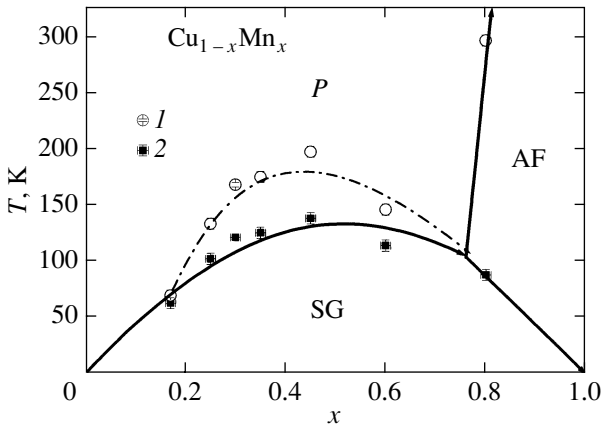

Fig. 5. Phase diagram of the homogeneous copper–manganese alloys $Cu_{1-x}Mn_x$.

It should be noted that the static-field parameter Δ can be obtained by processing the experimental data only in the case where $\Delta > \lambda_D$. Note also that a decrease in the parameter λ_D leads to an increase in the reliability for determining the static-field parameter Δ.

Therefore, the results obtained demonstrate that, in the homogeneous alloys $Cu_{1-x}Mn_x$ over a wide range of concentrations, there exists a phase transition to a specific magnetic state at temperatures in the range 100–200 K. This phase arises irrespective of the type of the high-temperature state, i.e., the paramagnetic or antiferromagnetic state. The new phase is characterized by a considerable nonuniformity of local fields due to the absence of a long-range magnetic order.

3. DISCUSSION AND CONCLUSIONS

Thus, the data obtained make it possible to complement substantially the magnetic phase diagram of the homogeneous copper–manganese alloys $Cu_{1-x}Mn_x$ (Fig. 5). This phase diagram takes the form characteristic of systems with competing exchange interactions [5].

In the phase diagram depicted in Fig. 5, the solid line indicates the boundaries between the paramagnetic (P), antiferromagnetic (AF), and spin-glass (SG) states according to the data available in the literature [1, 2, 4, 7]. Points *1* and *2* correspond to the results obtained in the present work for the high- and low-temperature transitions, respectively. The dot-dashed line represents the conventional boundary of the existence of the new phase state between the paramagnetic and spin-glass phases. As can be seen from Fig. 5, the concentration dependence $T(x)$ does not contradict the tendency of the change in the temperature T_G of the transition to the spin-glass state for concentrations $x < 0.15$. The largest temperature range between two transitions is observed at concentrations $x \sim 0.5$. The experimental results indicate that the new phase is characterized by a fast spin dynamics not only in the vicinity of the transition but also at lower temperatures up to the temperature of the transition to the spin-glass state. The analysis of the experimental data obtained allows us to assume that two magnetically ordered phases can be formed in the $Cu_{1-x}Mn_x$ binary alloys at relatively high concentrations of magnetic manganese atoms in the temperature range from 250 to 20 K. At higher temperatures (above 100 K), there arises a state with a fast spin dynamics and fluctuating random fields. In this state, the parameters λ_D and Δ are of the same order of magnitude. The experimental data obtained can be described using complex relaxation functions G [relationships (3), (4)]. At a temperature of ~70 K, the transition to the spin-glass phase without fluctuating random fields, i.e., the conventional spin-glass phase, is observed in the alloys for all the concentrations under investigation.

ACKNOWLEDGMENTS

This study was supported by the Branch of General Physics and Astronomy of the Russian Academy of Sciences.

REFERENCES

1. E. Z. Vintaĭkin, V. A. Udovenko, D. F. Litvin, S. Yu. Makushev, and V. B. Dmitriev, Izv. Vyssh. Uchebn. Zaved., Fiz. **5**, 104 (1985).
2. E. Z. Vintaĭkin, V. A. Udovenko, and V. B. Dmitriev, Fiz. Met. Metalloved. **44**, 1023 (1977).
3. A. N. Vasil'ev, V. D. Buchel'nikov, T. Takagi, V. V. Khovaĭlo, and É. I. Éstrin, Usp. Fiz. Nauk **173** (6), 577 (2003) [Phys. Usp. **46** (6), 559 (2003)].
4. A. Banerjee and A. K. Majumdar, Phys. Rev. B: Condens. Matter **46**, 8958 (1992).
5. S. G. Barsov, A. L. Getalov, S. L. Ginsburg, V. P. Koptev, S. P. Kruglov, L. A. Kuzmin, S. V. Maleyev, E. I. Maltsev, 1 S. M. Mikirtychyans, N. A. Tarasov, G. V. Shcherbakov, V. G. Grebinnik, V. N. Duginov, A. B. Lazarev, V. G. Olshevski, S. N. Shilov, V. A. Zhukov, I. I. Gurevich, B. F. Kirillov, A. I. Klimov, B. A. Nikolski, A. V. Pirogov, A. N. Ponomarev, and V. A. Suetin, Hyperfine Interact. **64**, 415 (1990).
6. S. G. Barsov, G. P. Gasnikova, A. L. Getalov, V. P. Koptev, S. A. Kotov, L. A. Kuz'min, A. Z. Men'shikov, S. M. Mikirtych'yants, and G. V. Shcherbakov, 2 Pis'ma Zh. Éksp. Teor. Fiz. **57** (10), 651 (1993) [JETP Lett. **57** (10), 672 (1992)].
7. Y. J. Uemura, T. Yamazaki, D. R. Harshman, M. Semba, and E. J. Ansoldo, Phys. Rev. B: Condens. Matter **31**, 546 (1985).
8. S. G. Barsov, A. L. Getalov, V. P. Koptev, S. A. Kotov, S. M. Mikirtychyans, and G. V. Shcherbakov, Physica B (Amsterdam) **289–290**, 221 (2000).
9. S. G. Barsov, F. L. Getalov, V. P. Koptev, L. A. Kuz'min, S. M. Mikirtych'yants, N. A. Tarasov, and G. V. Shcher- 2 bakov, Preprint No. 1312 LIYaF (Leningrad Institute of Nuclear Physics, Gatchina, Leningrad Region, 1988) [in Russian].

Translated by O. Borovik-Romanova

SPELL: 1. Maleyev, 2. Mikirtych'yants or Mikirtychyans?