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 $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$ with frustrated magnetic structure

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2001 J. Phys.: Condens. Matter 13 L127

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LETTER TO THE EDITOR

The behaviour of the magnetostriction and magnetoresistance of the ferrite $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$ with frustrated magnetic structure

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Received 29 November 2000

Abstract

The behaviour of the spontaneous magnetization σ_s , coercive force H_c , magnetostriction λ , magnetoresistance $\Delta R/R$ and electroresistance R of the ferrite spinel $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$ with frustrated magnetic structure has been investigated.

It was found that for a given sample the anomalous behaviour of the magnetostriction and magnetoresistance that occurs differs from that of usual ferrimagnetic structures, in which the long-range magnetic order penetrates the whole volume of a sample. For example, both the parallel (λ_{\parallel}) and the perpendicular (λ_{\perp}) magnetostriction throughout the interval of temperatures investigated in all fields have only positive sign.

It has been also established that there is anomalous behaviour of the magnetoresistance: both the parallel ($(\Delta R/R)_{\parallel}$) and the perpendicular ($(\Delta R/R)_{\perp}$) magnetoresistance, which are negative throughout the interval of temperatures investigated, decrease monotonically with increasing temperature.

The sample investigated shows two magnetic phase transitions when the temperature decreases. The first transition, at the Curie temperature $T_C = 460 \pm 5$ K, occurs from the paramagnetic state to a phase which consists of spontaneously magnetized areas formed due to short-range order (clusters). The second transition, from this phase to a state with frustrated magnetic structure, takes place at a lower temperature, $T_f = 410 \pm 5$ K.

1. Introduction

The frustrated magnetic structure in ferrite with the spinel structure can arise when there is replacement of magnetic ions by nonmagnetic ones [1, 2]. In this case the frustrated magnetic structure consists of separate spontaneously magnetized areas formed due to long-range and short-range magnetic order.

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In such magnetic structures, dilution induces a decrease (in absolute value) of the average AB interaction with respect AA and BB ones. As a consequence, the competition between negative intrasublattice and intersublattice interactions becomes strong, which leads to the formation of a cluster spin-glass state and frustrated magnetic structure [3].

In view of this, it is of interest to investigate the behaviours of the magnetostriction and of the magnetoresistance of a sample with frustrated magnetic structure and to carry out a comparison with the behaviours of these properties for samples with the usual nonfrustrated magnetic structures.

2. Experiment

The sample $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$ was prepared using ceramic technology. A first anneal was carried out at a temperature of 750 °C for 20 h; a second one was performed at a temperature of 900 °C for 20 h with subsequent slow cooling. Both firings were carried out in air. X-ray diffraction patterns recorded at room temperature indicated that the sample was single-phase spinel.

The measurements of the magnetization σ and coercive force H_c were carried out by the ballistic method in magnetic fields up to 10 kOe in the temperature range 80–650 K. The magnetization was measured for a ball with a diameter of 7 mm. The residual magnetization σ_r and coercive force H_c were derived from the shapes of the hysteresis loops.

The magnetostriction was measured by means of tensiometers in magnetic fields up to 12 kOe in the temperature range 80–400 K. A sample of dimensions $7 \times 7 \times 2 \text{ mm}^3$ was used for the magnetostriction measurement. The magnetoresistance and electroresistance were measured by a bridge method using contacts made with silver paste in the temperature range 293–500 K in fields up to 12 kOe.

The relative errors in the magnetization, magnetostriction and electroresistance measurements were about 3%. The relative errors in measuring the magnetoresistance did not exceed 5%.

3. Results and discussion

The curves for $\sigma_s(T)$, $H_c(T)$ and $(d\sigma_s/dT)(T)$ for $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$ are represented in figure 1. For this ferrite there are anomalous behaviours of the spontaneous magnetization $\sigma_s(T)$ and coercive force $H_c(T)$: abrupt decrease in the value of σ_s occurs at temperatures lower than those at which the value of H_c drops. One can see that at the temperature $T_i = 410 \pm 5 \text{ K}$ which is found by extrapolating the linear part of the curve $\sigma_s(T)$ to the temperature axis, the coercive force H_c remains rather high and vanishes at the higher temperature $T_C = 460 \pm 5 \text{ K}$. So, the Curie temperature T_C of this sample was defined as the temperature at which both the magnetization σ_s and coercive force H_c vanish. The measurements of the residual magnetization σ_r for the sample $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$ show that throughout the whole temperature range investigated, σ_r does not change its sign; i.e. the observed abrupt fall in the value of σ_s at $T = T_i$ is not connected with the compensation temperature, but has another cause [4].

It is established that the behaviour of the derivative $d\sigma_s/dT$ is anomalous: in the region of the Curie temperature T_C in the curve for $d\sigma_s/dT$ there is no maximum, whereas for a usual ferrimagnet a maximum should occur. Moreover, for this sample the curve $\sigma_s(T)$ is an almost linear function of the temperature over rather a large temperature range.

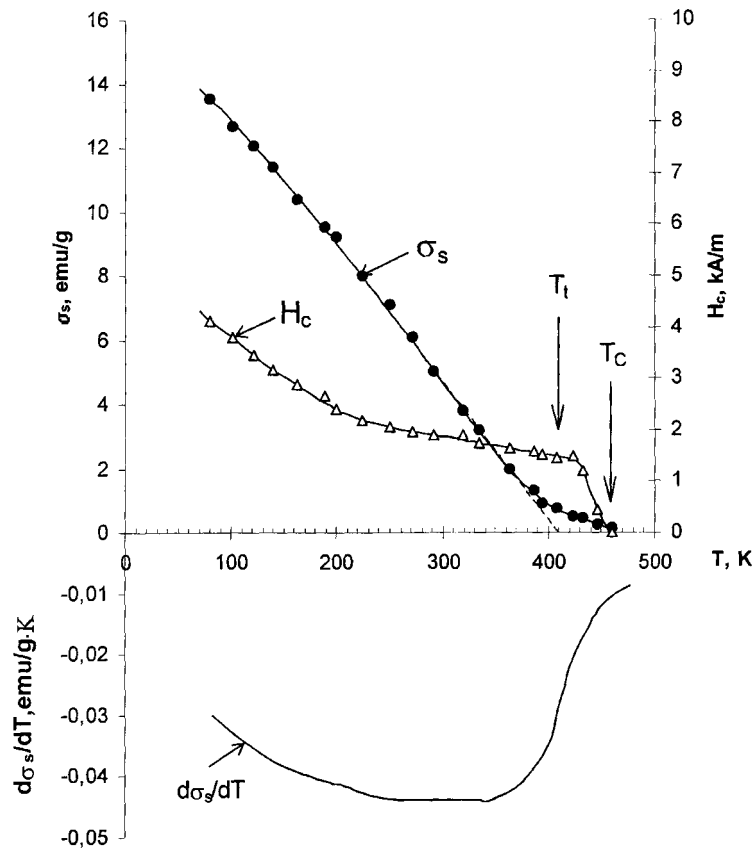


Figure 1. Temperature dependences of the spontaneous magnetization σ_s , coercive force H_c and derivative $d\sigma_s/dT$ for the sample of $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$.

We investigated the magnetization isotherms for the ferrite $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$ by the method of thermodynamic coefficients [5]; i.e. the curves for $(H/\sigma)(\sigma^2)$ were drawn. It turned out that a long-range magnetic order appears at temperatures lower than the temperature $T \approx 413$ K which for the given sample is approximately equal to T_t .

The magnetization results obtained also conform with the conclusions from the theoretical model of Van Hemmen [6], which indicates that if in a magnetic composition there is a transition from a paramagnetic to a spin-glass state, then at still lower temperatures a second magnetic phase transition from the spin-glass state to a frustrated magnetic phase must emerge. In frustrated magnetic phases there are fairly large spontaneously magnetized areas formed due to long-range magnetic order, which are chaotically arranged over whole volume of the sample.

Electroresistance measurement shows that for this ferrite the activation energy change occurs at two temperatures: first, a jump of the activation energy from 0.34 eV to 0.38 eV takes place at a temperature of about 413 K, and then a second one from 0.38 eV to 0.47 eV occurs at a temperature of about 460 K. So it turned out that the temperatures at which the activation energy jumps take place are close to the transition temperatures T_t and T_C .

It is well known that in the case of frustrated magnetic structures, there are large, separated, spontaneously magnetized areas formed due to long-range magnetic order. In this case, the value of the magnetization, which depends on the magnetic field, is governed by the rotation of

the magnetic moments of these areas with respect to the field. In these structures, the technical magnetism must be practically zero, and mostly paramagnetic processes will be observed [7, 8]. In this case, the sample will be total magnetized owing to rotation of the magnetic moments of the separated clusters with respect to the direction of the field.

Hence for the determination of the type of the magnetic structure, measurements of the parallel λ_{\parallel} and perpendicular λ_{\perp} magnetostrictions are necessary, because in the technical magnetism region they have anisotropic behaviour, whereas they are isotropic in the region of paramagnetism.

The isotherms of the magnetization σ , and the parallel (λ_{\parallel}) and perpendicular (λ_{\perp}) magnetostrictions, measured at $T = 86.5$ K, are represented in figure 2. One can see that for the curve $\sigma(H)$ there is no saturation. The magnetostrictions λ_{\parallel} and λ_{\perp} in all fields have isotropic character. Moreover λ_{\parallel} and λ_{\perp} are already equal to each other in small magnetic fields throughout almost the whole temperature range under study; i.e. the role of technical magnetism in these ferrites is insignificant, and paramagnetism is mostly observed [4].

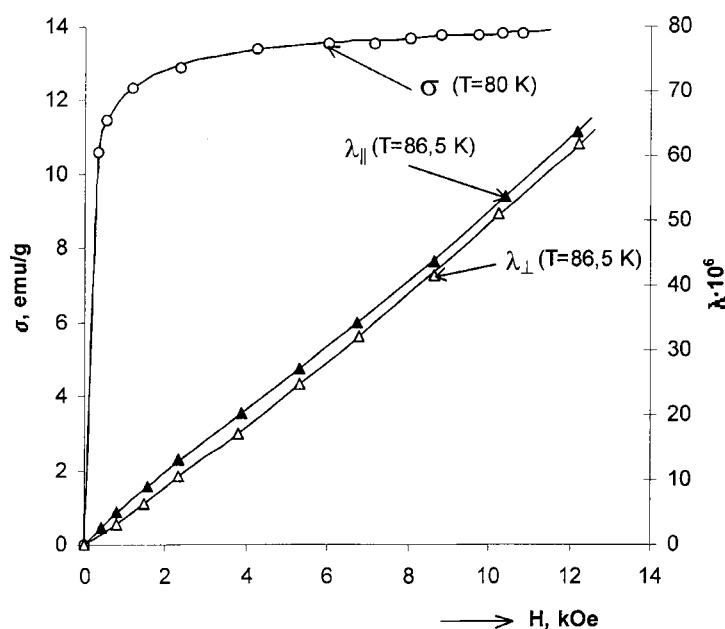


Figure 2. Isotherms of the magnetization (σ , for $T = 80$ K) and parallel (λ_{\parallel}) and perpendicular (λ_{\perp}) magnetostrictions (for $T = 86.5$ K) for the sample of $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$.

Figure 3 shows the temperature dependencies of the magnetostrictions λ_{\parallel} and λ_{\perp} , measured in a magnetic field of $H = 12$ kOe. The temperature dependence of the spontaneous magnetization is given in figure 3 by a dotted line. It can be seen that the magnetostrictions λ_{\parallel} and λ_{\perp} are positive and have the same value over a wide temperature range. For usual ferrimagnets with nonfrustrated magnetic structure in the technical magnetic regions, as a rule λ_{\parallel} and λ_{\perp} have opposite signs and obey the relation $\lambda_{\parallel} = -2\lambda_{\perp}$. The temperature dependences of the volume (ω) and anisotropic (λ_r) magnetostrictions in a field of $H = 12$ kOe are also represented in figure 3, where ω and λ_r have been calculated using the formulae $\omega = \lambda_{\parallel} + 2\lambda_{\perp}$ and $\lambda_r = \lambda_{\parallel} - \lambda_{\perp}$. One can see that the anisotropic magnetostriction λ_r is very small, whereas the volume magnetostriction ω is high and has a positive sign. It should be noted that the magnetostrictions become practically zero on approach to T_f .

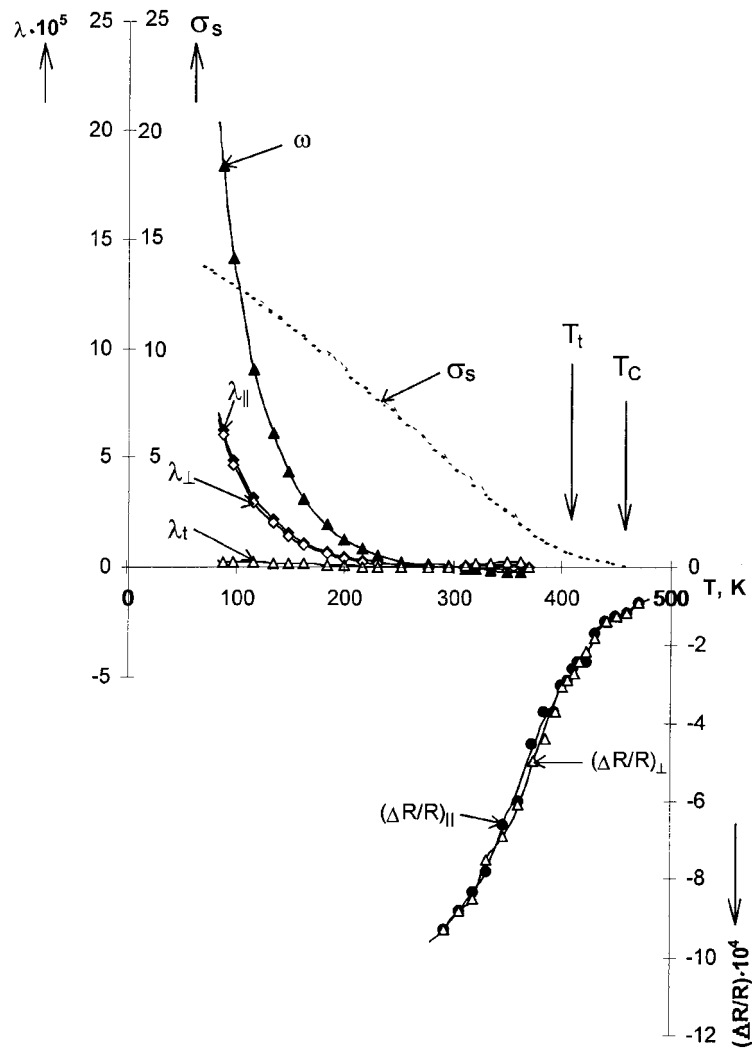


Figure 3. Temperature dependences of the parallel (λ_{\parallel}), perpendicular (λ_{\perp}), volume (ω) and anisotropic (λ_t) magnetostrictions ($H = 12$ kOe); and the parallel ($(\Delta R/R)_{\parallel}$) and perpendicular ($(\Delta R/R)_{\perp}$) magnetoresistances ($H = 12$ kOe); the temperature dependence $\sigma_s(T)$ is given by a dotted line for the sample of $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$.

Figure 3 also shows the temperature dependencies of the parallel ($(\Delta R/R)_{\parallel}$) and perpendicular ($(\Delta R/R)_{\perp}$) magnetoresistances in a field of $H = 12$ kOe. Unfortunately, below room temperature we could not measure $\Delta R/R$, as the resistance ($R \approx 3.45 \times 10^6 \Omega$) has an anomalously large value. It can be seen that in the neighbourhood of the temperature T_t , where the abrupt fall in the spontaneous magnetization occurs, extrema in the temperature dependencies of the magnetoresistances $(\Delta R/R)_{\parallel}$ and $(\Delta R/R)_{\perp}$ are absent. It should be noted that the extrema of the curves $(\Delta R/R)_{\parallel}(T)$ and $(\Delta R/R)_{\perp}(T)$ are absent also at the temperature T_C at which the coercive force goes to zero. For a usual ferrimagnet the maximum in the temperature dependencies of the magnetoresistance should be observed at the temperature of formation of the long-range magnetic order on cooling the sample.

The isotherms of the parallel magnetoresistance $(\Delta R/R)_{\parallel}(H)$ at various temperatures are shown in a figure 4. It should be noted that the behaviour of the isotherms for the perpendicular magnetoresistance $(\Delta R/R)_{\perp}(H)$ is similar to the behaviour of the curves for $(\Delta R/R)_{\parallel}(H)$. It is apparent that the paramagnetic regions of the $(\Delta R/R)_{\parallel}(H)$ isotherms only show a monotonic increase with the reduction of temperature throughout the interval of temperatures investigated, whereas for usual ferrimagnets the paramagnetic regions of the isotherms of the magnetoresistance should have their highest values in the neighbourhood of the Curie point.

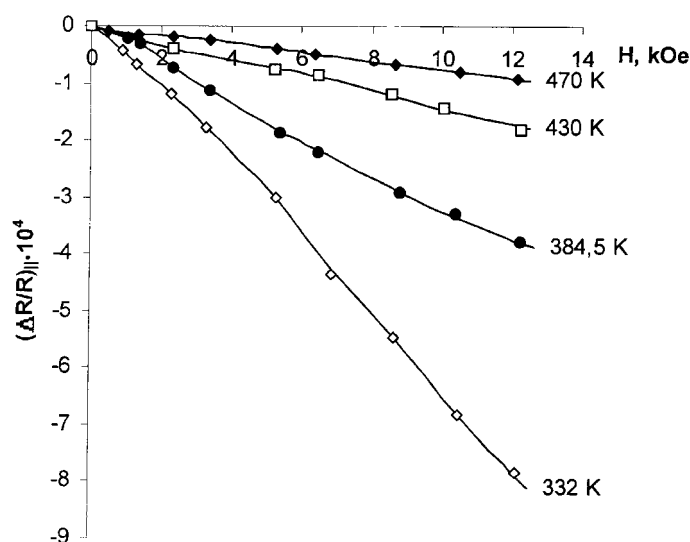


Figure 4. Isotherms of parallel magnetoresistance $((\Delta R/R)_{\parallel}(H))$ for the $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$ sample.

From the behaviour of the magnetoresistances $(\Delta R/R)_{\parallel}$, $(\Delta R/R)_{\perp}$ and the coercive force $H_c(T)$, one can suspect that there is a frustrated magnetic structure at temperatures lower than T_f . The negative sign of the two magnetoresistances at $H = 12$ kOe and their approximate equality indicate that the sample behaviour in strong magnetic fields can be described in terms of paramagnetic processes.

The anomalous behaviour of the magnetostrictions for the ferrite $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$ —i.e. the presence of a large magnetostriction for paramagnetism whereas magnetostriction for technical magnetism is practically absent—confirms our supposition of the formation of frustrated magnetic structure in this ferrite at temperatures lower than T_f . It is possible that the process of magnetization in this ferrite is due to the rotation of magnetic moments of spontaneously magnetized areas with respect to the magnetic field.

4. Conclusions

It is established that for the ferrite $\text{CuGa}_{0.4}\text{Al}_{0.8}\text{Fe}_{0.8}\text{O}_4$ with frustrated magnetic structure, there is anomalous behaviour of a number of magnetic properties. The spontaneous magnetization $\sigma_s(T)$ depends linearly on temperature over a large temperature interval, and the reduction of the value of σ_s occurs at lower temperature than the reduction of the coercive force. The assumption is made that two magnetic phase transitions take place for the given ferrite: at T_f ,

the transition from the paramagnetic state to a spin-glass state; and at T_C , that from a spin-glass state to a frustrated magnetic phase.

It is revealed that near the temperatures T_i and T_C there is a change in the energy of activation. Anomalous behaviour of the magnetoresistance is revealed: with the increasing of temperature both the parallel ($(\Delta R/R)_{\parallel}(T)$) and the perpendicular ($(\Delta R/R)_{\perp}(T)$) magnetoresistance only decrease monotonically with increase of temperature and do not show a jump in magnetoresistance at T_i or at T_C . Thus the paramagnetic regions of the $(\Delta R/R)_{\parallel}(H)$ and $(\Delta R/R)_{\perp}(H)$ magnetoresistance isotherms only decrease monotonically with increase of temperature.

Anomalous behaviour of the magnetostriction is also revealed: the parallel ($\lambda_{\parallel}(T)$) and perpendicular ($\lambda_{\perp}(T)$) magnetostriction have identical values and positive sign throughout the interval of temperatures investigated. Thus, large paramagnetic regions are also observed at low temperatures in the curves for $\lambda_{\parallel}(H)$ and $\lambda_{\perp}(H)$. It is established that at low temperatures there is a large volume magnetostriction (ω), whereas the anisotropic magnetostriction (λ_t) is practically zero.

The author is very grateful to Dr A N Goryaga for the discussion of results.

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