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Anomaly in the Temperature Dependence of the Coercive Force of an Ensemble of Ferromagnetic CrO_2 Nanoparticles under Conditions of Percolation Conductivity

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Abstract—The temperature dependence of coercive force $H_c(T)$ of pressed powders of ferromagnetic half metal CrO_2 is compared to the temperature dependence of the maximum field of positive tunneling magnetoresistance $H_p(T)$. It is established that the multidomain alters the nature of the reverse magnetization of transport channels and violates relation $H_p \approx H_c$ at low temperatures (4.2–70 K).

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INTRODUCTION

Chromium dioxide (CrO_2) is known as half metal I_A, which has only electrons with up spins (↑) at the Fermi level (E_F). Pressed chromium dioxide powders are a cooperative system of magnetic tunnel contacts with a percolation conductivity at low temperatures. Since chromium dioxide is widely used in the production of hard disks with high information recording density, the problem of increasing the coercive force of CrO_2 powder is of great interest to researchers and technologists. The stronger the coercive force, the higher the material's capability to resist demagnetizing fields. The maximum value of the coercive force is characteristic of a magnetic material consisting of single-domain particles. The production of powders with high coercive force is reduced in particular to the problem of synthesizing single-domain chromium dioxide nanoparticles. This problem can be solved by controlling the size and shape of nanoparticles using small amounts of modifying additives during hydrothermal synthesis [1].

On the other hand, some granules are in the multidomain state even in a system of single-domain particles. A multidomain state arises due to negligible dispersion in granule sizes, and to weak tunnel barriers between some granules. The low level of dispersion in granule size and the thickness of the dielectric shells of the granules results in percolation conductivity, due to the existence of optimum chains of granules with the maximum probability of tunneling for adjacent pairs that form a conducting chain [2]. The number of conducting chains falls continuously along with temperature under conditions of activated conductivity, and the percolation grid can even be reduced to one conducting channel at a sufficiently low temperature [2]. These optimum chains inevitably have a number of weak links (high-resistance tunnel junctions) with increased activation energy that determine the activated character of the total measured conductivity. The configuration of an infinite conducting cluster gradually changes as its density falls along with temperature. These phenomena alter the ratio between the number of multidomain particles that participate in conductivity and the total number of single-domain particles, most of which do not participate in charge transfer at low temperatures. The multidomain nature of the granules of an infinite conducting cluster will influence its magnetic properties and the character of the temperature dependence of the coercive force of transport channels.

The aim of this work was to detect and analyze the possible difference in the coercive force of the macroscopic system of ferromagnetic (FM) granules from the coercive force of transport channels that form the percolation grid at low temperatures.

EXPERIMENTAL

Coercive force H_c of a macroscopic system was determined from the hysteresis measurements of the global magnetization of pressed powders at a fixed temperature. The value of H_c was determined from the anisotropy fields blocking the process of reverse magnetization. Coercive force H_p of the system of FM

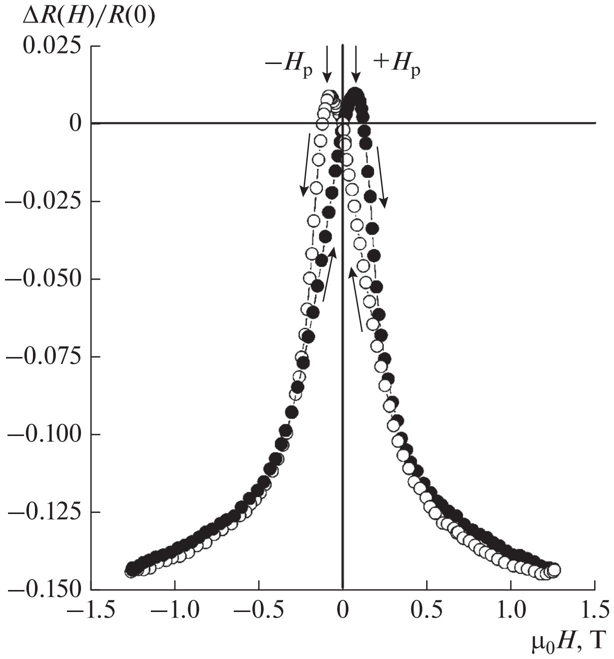

Fig. 1. MR hysteresis of pressed powder CrO_2 (powder no. 1) at $T = 30.03$ K. The arrows indicate the direction of change of the magnetic field when recording the MR curves. Field H_p corresponds to the magnetic field in which the maximum value of the resistance $R(H)$ is reached.

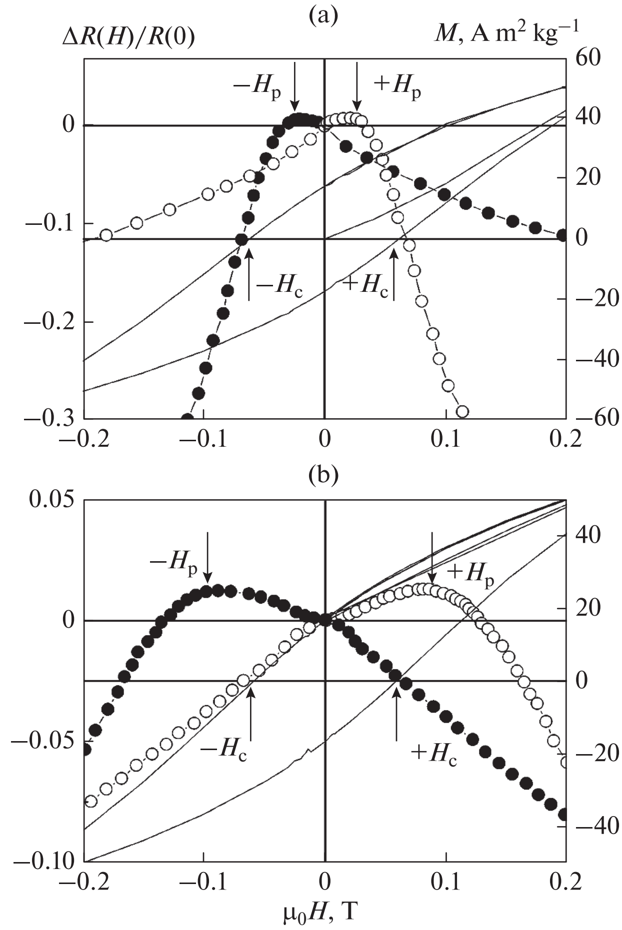

Fig. 2. MR hysteresis (left y-axis) and magnetization hysteresis (right y-axis, upper x-axis) of sample no. 2 at temperatures of (a) 5 K and (b) 20 K in the region of small fields.

granules that form the transport channels corresponded to the maximum field of positive magnetoresistance (MR) (Fig. 1). The value of H_p was determined from the transport measurements of MR hysteresis.

In this work, we considered the dependence of the coercive force on temperature for several pressed chromium dioxide powders consisting of nanoparticles coated with dielectric shells with different types and thicknesses. The main characteristics of the investigated powders are given in Table 1.

A strong difference between the H_p and H_c values in the low-temperature region was noted in [3, 4]. This difference is clearly seen in Fig. 2, where the regions of hysteresis of tunneling MR and magnetization in the areas of small fields are shown for powder no. 2. It is seen that $H_p < H_c$ at $T = 5$ K, and $H_p > H_c$ at $T = 20$ K for this powder. Thus, relation $H_p \approx H_c$ for sample no. 2 is not satisfied at low temperatures.

Figure 3 shows the temperature dependences of H_c and H_p, recorded in the temperature range of 5–300 K

Table 1

Sample, no.	Powder composition	Particle shape	Type of dielectric shell	Shell thickness, nm	Particle length, nm	Particle diameter, nm	H_c, Oe (room temperature)
1	CrO_2	Needles	β-CrOOH	1.73	302	22.9 ± 0.8	432
2	CrO_2	Needles	Cr_2O_3	2.1	302	22.9 ± 0.8	422
3	CrO_2	Spherical	β-CrOOH	3.6	–	120	149
4	CrO_2	Needles	β-CrOOH	1.58	302	22.9 ± 0.8	429

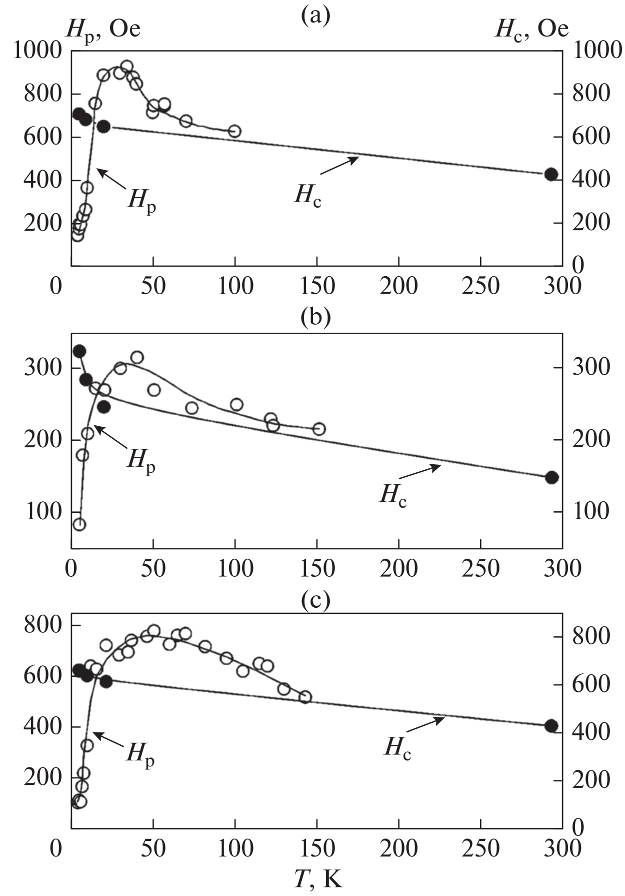

Fig. 3. Temperature dependences of field H_p (the peak of the positive MR) and coercive force H_c (left y-axis, upper x-axis) of three different samples ((a) sample no. 1, (b) sample no. 3, and (c) sample no. 4).

for samples 1, 3, and 4. The value of H_c for ferromagnets is normally maximal at low temperatures and falls as the temperature rises (in the limit to zero when approaching Curie temperature T_C). Dependence $H_c(T)$ in Fig. 3 corresponds to this behavior. At the same time, dependence $H_p(T)$ is unusual. $H_p(T)$ behavior is characterized by relation $H_p \approx H_c$ not being satisfied for samples nos. 1, 3, and 4; or for sample no. 2, even though this was expected and observed for pressed powders with fairly small (submicron) sizes, including CrO_2 powders [5, 6]. Dependence $H_p(T)$ is nonmonotonic as well. The value of H_p considerably exceeds H_c in the 50–100 K region of temperatures, and the difference between H_p and H_c is reduced considerably upon a further increase in temperature. The sign of derivative dH_p/dT changes in the low-temperature region ($T < 50$ K), and H_p falls sharply along with temperature. Ratio H_p/H_c diminishes rapidly along with temperature in the temperature range of $T \leq 20$ K.

The optimum conducting chains of granules that make the main contribution to conductivity consist mainly of multidomain particles. Local values H_{c1} of these multidomain particles are considerably lower than the H_c of single-domain particles ($H_{c1} < H_c$), so the relation $H_p < H_c$, where H_p is the coercive force of an infinite conducting cluster, must be satisfied at low temperatures. Transport channels gradually freeze as the temperature falls. Clusters in which the number of multidomain particles is larger survive in this process. This apparently leads to a further reduction in the H_p cluster as the temperature falls.

At the same time, nonmonotonic dependence $H_p(T)$ (there is a maximum of H_p) found in this work and the satisfaction of the relation $H_p > H_c$ in the region of relatively low temperatures $T \approx (30–50)$ K remain largely unclear. Higher H_p values have been observed in for $La_{2/3}Sr_{1/3}MnO_3$ manganite powders consisting of multidomain particles [7]. Study of five samples with different grain sizes showed [7], that the ratio $H_p \neq H_c$ is performed for multidomain particles of manganites. In addition, the value of H_p exceeds the value of H_c and it gradually increases with increasing grain size. The value of H_p tends to H_c as the particle size shrinks. When the particle size approaches the critical size of the single-domain state, relation $H_p \approx H_c$ begins to hold. The authors of [7] suggested that higher values of H_p relative to H_c were associated with features of magnetization reversal of the volume of multidomain grains and the regions close to grain boundaries. When most of the domains inside a grain volume switch, domains close to the interface between adjacent granules remain disoriented, making a notable contribution to conductivity that does not depend on the measured total magnetic moment.

Despite our level of understanding, we believe that many aspects of the problem of the H_p and H_c relationship are still unclear and require further research.

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