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THE HALL EFFECT IN FERRITES

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1. INTRODUCTION

FERRITES are ferrimagnetic semiconducting materials of the oxide group.* Despite the fact that ferrites have found wide and varied applications in technology, there are so far no sufficiently complete ideas about the nature of their magnetic and electric properties. Especially insufficient is the information about the nature of the electrical properties of ferrites. There is no clear understanding of the mechanism of electrical conduction in ferrites; the effect of their ferrimagnetic state on the electrical properties remains to be investigated, as does the effect of different forms of scattering of the current carriers on the galvano- and thermomagnetic phenomena, among them the anomalous Hall effect.

In a considerable portion of the experimental work on study of the electrical properties of ferrites, all that was measured was their electrical conductivity and thermoelectromotive force. It is well known, however, that the most complete information about the mechanism of conduction can be obtained from data on the Hall effect; these make possible a quantitative determination of the change in the number of carriers and in their mobility with temperature. Meanwhile, a very small number of researches have been devoted to investigation of the Hall effect in ferrites. In contrast, the Hall effect in metallic ferromagnets has been the object of intensified investigation over a period of many years.

These investigations have been developed especially successfully during recent years in the works of Soviet physicists[1-17]. In these works, and also in the works of foreign authors[18-27], there has been success in establishing the reasons for the occurrence in metallic ferromagnets of an anomalous Hall effect, and the character of the dependence of the Hall coefficients on the temperature and on the electrical resistivity.

The laws deduced in the investigation of the Hall effect for metals can also be carried over, to a certain degree, to ferrites. In this case, however, it is necessary to take into account their semiconductor mechanism of conduction and their ferrimagnetic type of magnetic order, which introduce special peculiarities into the Hall effect properties.

In the present article, an attempt is made to systematize the existing results (mainly experimental) on the Hall effect in ferrites. Considerable space is devoted to analysis of experimental data obtained by the authors of this review.

On the basis of the results of measurement of the Hall effect in ferrites, the so-called "jump" mechanism of conduction in ferrites (Verwey's hypothesis) is considered critically. This hypothesis has been widely used, up to now, by investigators in their study of the electrical properties of ferrites.

2. THE HALL EFFECT IN MAGNETICALLY ORDERED MATERIALS. METHODS OF DETERMINING THE CLASSICAL AND SPONTANEOUS HALL COEFFICIENTS

For substances that possess magnetic ordering (ferro-, ferri-, and antiferromagnets), the Hall electromagnetic force E, referred to unit current density, can be expressed as the sum of the classical and magnetic Hall fields[28-34]:

$$E = R_0 H + R_s I, \tag{1}$$

where I is the magnetization, R_0 is the classical Hall coefficient (dependent on the number of current carriers), and R_S is the spontaneous, or anomalous, Hall coefficient.* It is called anomalous because the numerical value of this coefficient is two or three orders of magnitude larger than the coefficient R_0. In paramagnets, the coefficient $R_S = R_p$ is called the paramagnetic Hall coefficient[1,31,32]. Most measurements of the Hall emf, both in metallic ferromagnets and in ferrites, have been made at constant current[28-36]. Although the Hall emf in ferrites is three to four orders of magnitude larger than the Hall emf in metallic ferromagnets, the technique for measurement of the Hall effect in ferrites is more difficult. The difficulties are basically due to the instability of the zero in measurement of the Hall emf by a potentiometric method. The quality of the electrical contacts has an important influence on the stability of the zero. The contacts to the surfaces of the ferrite are made in most cases by the method of brazing with silver paste. As is well known, on the surface between a semiconductor and a metal there are fluctuational emfs, which depend on the specific structure of the contacts[37]. It is possible that the instability of the zero depends also on the "noise" of the magnetized ferrite[38].

The current through the ferrite specimen is chosen experimentally, depending on the purity of the specimens and the quality of the contacts; it usually varies over the range 1 to 100 mA. In investigation of the Hall effect in ferromagnets, it is convenient to use the method of the "sliced" specimen, proposed in[42]. Figure 1 shows schematically the form of a "sliced"

* At present other semiconducting materials are also known that possess ferri- and antiferromagnetic properties, for example the group of compounds of the rare-earth metals with selenium and tellurium.

* Since the deflection of the electrons under the influence of the Lorentz force is proportional to the magnetic induction $B = H + 4\pi I$, in a number of papers [7,11,13] the relation (1) is written in the form $E = R_0 B + 4\pi R_S I$. In view of the fact that $R_S \gg R_0$, it is permissible to use relation (1), which is much more convenient.

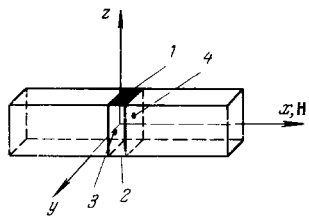

FIG. 1. "Sliced" specimen for measurement of the Hall effect in ferrites.

ferrite specimen. The vector magnetic field intensity H is directed along the specimen (the x axis). The demagnetizing factor of the specimen is smallest in this case. The electric current is fed to the silvered surfaces 1 and 2 of the parallelepiped, in the z direction. The Hall emf being measured is directed along the y axis (contacts 3 and 4). Into slits on each side of the parallelepiped are inserted thin wafers of mica, and thus all three parts of the specimen are isolated from one another. This method makes it possible to determine the current density in the specimen most precisely and to obtain a large value of it by use of comparatively small voltages. This method is especially convenient in measurement of the Hall emf on ferrites with a large electrical resistance.

The coefficients R_S and R_0 in most researches were determined as follows: the measured value of the Hall emf E is plotted as a function either of the magnetic field H (Fig. 2) or of the magnetic induction B. The slopes of the tangents to the E(H) curve at small and at large values of the magnetic field H are taken for the coefficients R_S and R_0 respectively.

In[36,43], attention was called to errors that may occur in this method of calculating the values of R_S and R_0 if the materials under study possess a noticeable paraprocess. These errors consist in the following: the increment ΔE in the Hall emf, corresponding to the paraprocess, can be written in the form[43]

$$\Delta E = R_0 \Delta H + R_s I_i, \qquad (2)$$

where $I_i = I - I_S$ is the paraprocess magnetization and $\Delta H = (H - H_S)$ (I_S and H_S are the magnetization and field, respectively, at technical saturation).

On introducing the paraprocess susceptibility χ_i, we have

$$\Delta E = (R_0 + R_s \chi_i) \Delta H. \qquad (3)$$

It is seen that the slope of the straight line E(H) in the paraprocess region, numerically equal to the ratio $\Delta E/\Delta H$, depends not only on R_0 but also on the product $R_S\chi_i$. Neglect of the term in the relation (3) is not always possible. Figures 3 and 4 give isothermal E(H) curves for a Cr-Te alloy[1] and for a monocrystal of cobalt ferrite $Co_{0.94}Fe_{2.06}O_4$[44]. It is seen that the slope of the tangents to these curves in the paraprocess region varies greatly with temperature and with field H not only in magnitude, but also in sign (compare temperatures 8° and 54°C in Fig. 3 and curves 1 and 5, at 80° and 368°C, in Fig. 4). At the same time it is known that in the temperature interval under consideration, the sign of the current carriers, for example in cobalt ferrite, does not change[44]. Consequently, for the materials cited the influence of the paraprocess on the course of the E(H) curve is large. Therefore determination of numerical values of the coefficient R_0 by the method illustrated in Fig. 2 is extremely difficult.

The influence of the paraprocess can, however, be taken into account in the determination of R_0. In fact, on differentiating relation (1) with respect to H we get

$$\frac{\partial E}{\partial H} = R_0 + \left(R_s \frac{\partial I}{\partial H} + I \frac{\partial R_s}{\partial H}\right)$$

or

$$\frac{\partial E}{\partial H} = R_0 + \left(R_s + H \frac{\partial R_s}{\partial H}\right) \frac{\partial I_i}{\partial H}. \qquad (4)$$

Here for H = 0, the coefficient R_S is equal to the

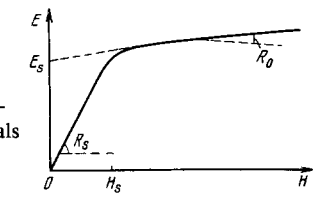

FIG. 2. Schematic curve of the dependence of the Hall emf on magnetic field H for ferromagnetic metals [101,109].

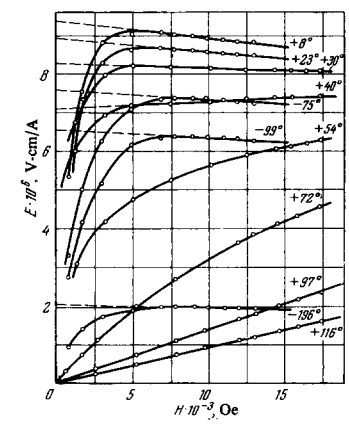

FIG. 3. Hall emf E as a function of the magnetic field H for a Cr-Te alloy.

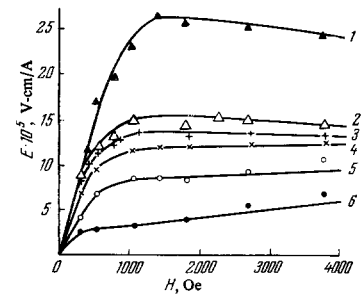

FIG. 4. Hall emf E as a function of magnetic field H for various temperatures in cobalt ferrite ($Co_{0.94}Fe_{2.06}O_4$). 1, 80°C; 2, 163°C; 3, 223°C; 4, 284°C; 5, 368°C; 6, 420°C.

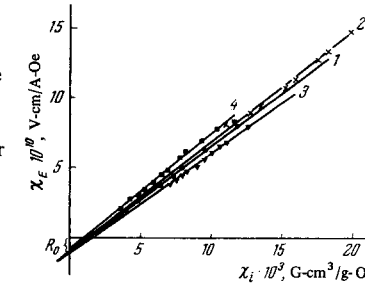

FIG. 5. Dependence of the Hall emf susceptibility χ_E on the paraprocess susceptibility χ_i for the alloy Fe_3Pt. 1,2, for hardened (50° and 82.5°C) specimen; 3, 163°C; 4, for annealed (182°C) specimen.

spontaneous coefficient, determined as the ratio E_S/I_S, where E_S and I_S are the spontaneous Hall emf and magnetization. The dependence of the Hall emf susceptibility $\partial E/\partial H = \chi_E$ on the paraprocess susceptibility $\partial I_i/\partial H = \chi_i$ is in general described by a curve whose intercept on the axis of ordinates is numerically equal to the classical Hall coefficient R_0. Figures 5 and 6 show this dependence for the magnetically ordered substances Fe_3Pt and $LeFe_2O_4$[47,49].

For the alloy Fe_3Pt, the value of R_0 is practically independent of temperature, whereas in the ferrite $LiFeO_4$, the intercept on the axis of ordinates decreases with increase of temperature; this indicates an increase of the number of current carriers. The slope of the curves in these figures, according to (4), is determined by the sum $R_S + H\partial R_S/\partial H$. If the derivative $\partial R_S/\partial H$ is small and the product $H\partial R_S/\partial H$ (the change of R_S in the paraprocess region) is negligible in comparison with the value of R_S, then these curves degenerate to the straight lines of Figs. 5 and 6.

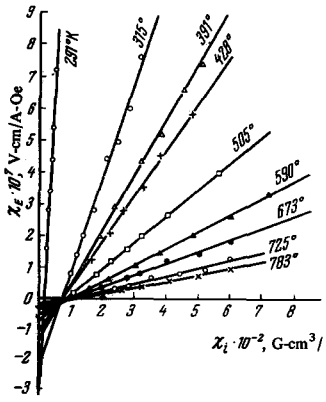

FIG. 6. Dependence of the Hall emf susceptibility χ_E on the paraprocess susceptibility χ_i for lithium ferrite $LiFe_2O_4$.

In[36] it was also shown that in the determination of the spontaneous Hall coefficient R_S, as the slope of the straight line E(H) or E(B) in the region of technical saturation (that is, by the method illustrated in Fig. 2), it is necessary in the neighborhood of the Curie temperature, where the spontaneous magnetization becomes small, to take account of the classical Hall field R_0H (in (1)). At small values of the magnetization, the first and second terms in relation (1) may be comparable in magnitude. Figure 7 shows two curves of the temperature dependence of the spontaneous Hall coefficient R_S for Fe_3Pt: curve 1 corresponds to the results of a calculation of R_S from the slope of the straight line E(H) at small H without allowance for the term R_0H, and curve 2 corresponds to values of R_S found from the slope of the straight line $(E - R_0H)$ as a function of the magnetization. It is seen that near the Curie temperature Θ, in the first case a maximum of the curve $R_S(T)$ was obtained, whereas in the second case the coefficient R_S increases monotonically to a value numerically equal to the paramagnetic Hall constant R_p; this is in agreement with the work of Kikoin. Consequently, the presence of a maximum on the $R_S(T)$ curve near the the Curie temperature, according to data of other authors[45,46], is caused by neglect of the term R_0H in relation (1) and does not correspond to the actual behavior of the $R_S(T)$ curve.

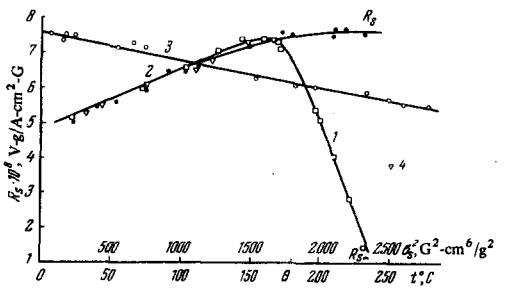

FIG. 7. Dependence of the spontaneous Hall coefficient R_S of the alloy Fe_3Pt on temperature for two different methods of calculating it: with allowance for the term R_0H (curve 2) and without allowance for the term R_0H (curve 1). Curve 3: spontaneous Hall coefficient as a function of the square of the spontaneous magnetization.

Recently Turov and Shavrov[114-116], starting from phenomenological considerations, have shown that in antiferromagnets and ferrimagnets there should be, in addition to the dependence of the Hall emf on H, I_S, and I_i, a dependence of E on the so-called antiferromagnetism vector L (this vector describes the direction of the axis of antiferromagnetism in the crystal and is numerically equal to the difference between the magnetization vectors of the sublattices: $L = I_{S1} - I_{S2}$). For ferrites, therefore, instead of relation (1) it is necessary to write in general

$$E = R_0H + R_sI + R_LL = R_0H + \left(R_s + R_L\frac{L}{I}\right)I. \qquad (5)$$

Thus the anomalous Hall constant due to the influence of the ferrimagnetic state of a ferrite on the Hall effect is

$$R_p = R_s + R_L\frac{L}{I}. \qquad (6)$$

It should be pointed out, however, that in most ferrites the second term in the relation (6) "does not show" itself. The fact is that the vector L, because of its varied orientation in a ferrite (especially polycrystalline), produces effects of the influence of the direction of L on the Hall emf which average out over the crystal and therefore are not observed. Only in ferrites that possess a compensation point Θ_C are there conditions favorable for observation of the influence of the direction of L on the Hall emf. On passage through Θ_C the direction of the vector L reverses. Therefore if $R_S < R_LL/I$, a change of sign of the Hall emf should be observed near Θ_C. This has been observed experimentally[117] in the alloy Mn_5Ge_2 ($\Theta_C = 122°C$). In this same alloy there has been discovered, near Θ_C, a change of sign of the galvanomagnetic effect $\Delta\rho/\rho$, which is also explained by the influence of a change of orientation of L[118].

For ferrites, measurements of the Hall effect in the compensation point region have not yet been made, but a change of sign of $\Delta\rho/\rho$ has been observed[119].

3. TEMPERATURE DEPENDENCE OF THE SPONTANEOUS HALL COEFFICIENT IN FERRITES

The presence of a spontaneous Hall field in ferro- and ferrimagnetic metals and semiconductors is attributed by contemporary theories[7,9-11,13,17] to the

fact that in them, besides scattering of the current carriers on impurities, defects, and thermal oscillations of the lattice, there is scattering on nonuniformities of the magnetic spin system. This leads to the appearance of a Hall field proportional to the spontaneous magnetization (see relation (1)). From the theory of the Hall effect it also follows that with change of temperature, the Hall coefficient R_S changes in proportion to the magnetic part of the electrical resistivity ρ_m, which appears through scattering of the current carriers on the spin system. Furthermore moment leads to the result that $\rho_m \neq 0$ even when $T \to 0°K$. This "magnetic impurity" part of the spontaneous Hall coefficient was recently determined[52] for an Invar alloy.

Experimental investigations of the Hall effect in metallic ferromagnets have established that the spontaneous Hall coefficient R_S is a linear function of the square of the spontaneous magnetization over a wide temperature interval[1,31-33,35,36,47,50-52,56]. By way of example, such relationships are shown in Fig. 7 (curve 3) and Figs. 8a and 8b. These relationships support the theories in which the coefficient R_S is proportional to the size of ρ_m. In fact, in metals the concentration of current carriers n is almost independent of temperature, while the "magnetic" mobility μ_m of the carriers, according to[9,10,3,51], is connected with the spontaneous magnetization by the relation

$$\frac{1}{\mu_m} = b\,(I_{s_0}^2 - I_s^2); \qquad (7)$$

here I_{S0} is the spontaneous magnetization at $T = 0°K$ and b is a coefficient of proportionality.

Above the Curie temperature, the magnetic part of the electrical resistivity reaches a maximum and is practically independent of temperature; consequently, the paramagnetic Hall coefficient R_p also becomes constant over a wide temperature interval. This is confirmed by experiment[1,33,34].

A theory of the Hall effect in ferrites was given in[9,10]. It was shown that in ferrites, just as in metallic ferromagnets, scattering of the current carriers on the spin system leads to a linear dependence of the coefficient R_S on ρ_m. In the case of ferrites, however, it is necessary to take account of the change with temperature of the number of carriers n. Consequently, the change of the size of R_S with temperature is determined by the temperature dependence of the concentration of carriers n and of their "magnetic" mobility μ_m. A typical curve of the change of the spontaneous Hall coefficient R_S as a function of the temperature is shown in Fig. 9.

If we eliminate the effect of the temperature dependence of the concentration n on the size of R_S, that is if we consider the ratio R_S/R_0 as a function of the temperature, then the dependence of this ratio on I_S^2 should be linear.

It has been shown[48] that the ratio R_S/R_0 in fact does vary linearly with I_S^2 over a wide temperature interval (100–600°K) for single-crystal manganese, nickel, and cobalt ferrites (Fig. 10, a and b). In Fig. 11 it is seen that the numerical value of R_S/R_0 increases with increase of temperature up to the Curie point. The coefficient R_S has a similar temperature behavior in metallic ferromagnets (compare with Fig. 7, curve 2).

It follows from the experimental results presented that in ferrites (as in metallic ferromagnets) the

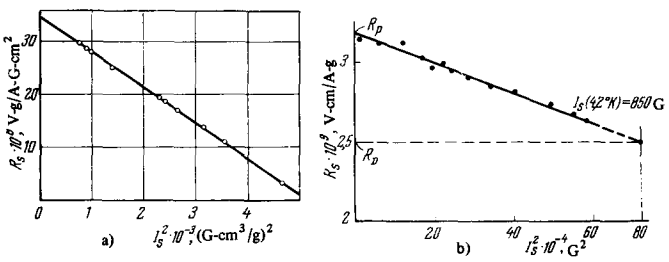

FIG. 8. Dependence of the spontaneous Hall coefficient on the square of the spontaneous magnetization: a) for a Cr-Te alloy; b) for an alloy of Invar type (62% Fe, 32% Ni, and 6% Cr).

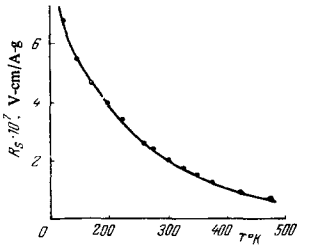

FIG. 9. Temperature dependence of spontaneous Hall coefficient for nickel ferrite ($Ni_{0.99}Fe_{2.31}O_4$).

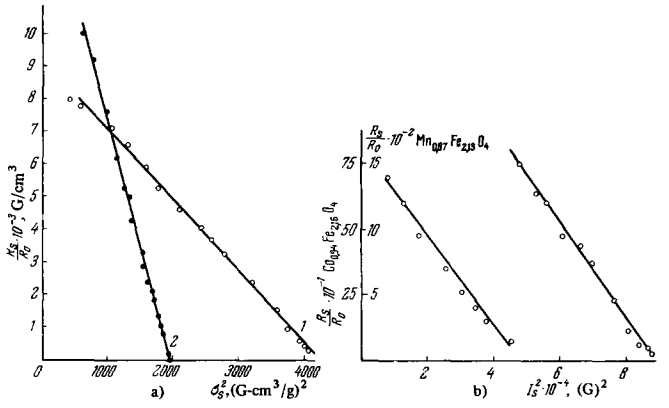

FIG. 10. Dependence of the ratio R_S/R_0 on the square of the spontaneous magnetization: a) for the ferrites $Ni_{0.69}Fe_{2.31}O_4$ (1) and $Mn_{0.76}Fe_{2.24}O_4$ (2); the scale for 1 is 10 times larger than the scale for 2; b) for Co and Mn ferrites; I_S is the spontaneous magnetization of unit volume (1 cm^3), σ_S is the spontaneous magnetization of unit mass (1 g).

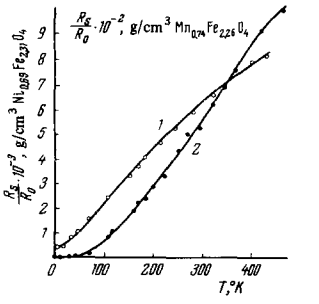

FIG. 11. Temperature dependence of the ratio R_S/R_0 for Mn ferrite (1) and Ni ferrite (2)

temperature dependence of the spontaneous Hall coefficient R_S is determined by the change of the magnetic resistivity ρ_m. This is perhaps the reason why the attempt of several authors[46,60] to establish a connection between the size of R_S and the total electrical resistivity ρ in ferrites, with change of temperature, has not given positive results.

4. THE HALL EFFECT IN THE PARAPROCESS REGION

In Sec. 3 we discussed the temperature dependence of the spontaneous Hall coefficient R_S in magnetic materials in the absence of an external magnetic field. Since for T = const the number of current carriers in ferrites is constant, the behavior of the Hall emf as a function of the magnetic field should be the same in metals and in ferrites. In the range of the technical magnetization curve, the coefficient R_S remains constant. This was first established for ferromagnetic metals in[30,32,29,41,53,54,65,110,111] and for ferrites in[58-60,102,112]. But study of the Hall effect in metals and in ferrites in the paraprocess region has shown that the spontaneous Hall coefficient R_S seemingly depends on the paraprocess magnetization. There have been attempts[14,43] to introduce a Hall constant R_i corresponding to the paraprocess magnetization I_i. It is more convenient, however, to consider a dependence of R_S on the paraprocess (that is, on the field H), since in this range the size of ρ_m as a rule decreases. Figure 12 shows curves of the Hall emf as a function of the magnetization for manganese and cobalt ferrites[46,59]. It is seen that in the paraprocess region, there occurs a departure from the linear dependence of E on I.

This departure might be ascribed to the influence of the term R_0H in Eq. (1), but allowance for this term does not lead to completely straightened out curves. In Fig. 13 it is seen that the slope of the curves of $(E - R_0H)$ as a function of the magnetization I in the paraprocess region, for the alloy Fe_3Pt and for a manganese ferrite, diminishes noticeably.

Theoretical and experimental consideration was given to this problem, for metals, in[3,13,51,66]. Since the spontaneous Hall coefficient R_S depends on the magnetic part ρ_m of the electrical resistivity, and since the size of ρ_m as a rule decreases in the paraprocess region, the decrease of the coefficient R_S in this region only confirms the deduction of the theory to the effect that the value of R_S depends on ρ_m.

There are also other experimental results, however, Figure 12c shows curves of the Hall emf as a function of the magnetization for an Invar alloy, which has a large paraprocess[50]. It is seen that after diminution of the value of R_S on the section AB, there begins a remarkable increase of the coefficient to a value R_i indicated on the figure as the slope of the straight line E(I) in the paraprocess region. The value of this constant is almost independent of temperature and numerically equal to the paramagnetic Hall constant. For explanation of such behavior of the Hall effect in the paraprocess region, complicated considerations of even and odd galvanomagnetic effects are necessary. Unfortunately, such researches are almost nonexistent[62,67].

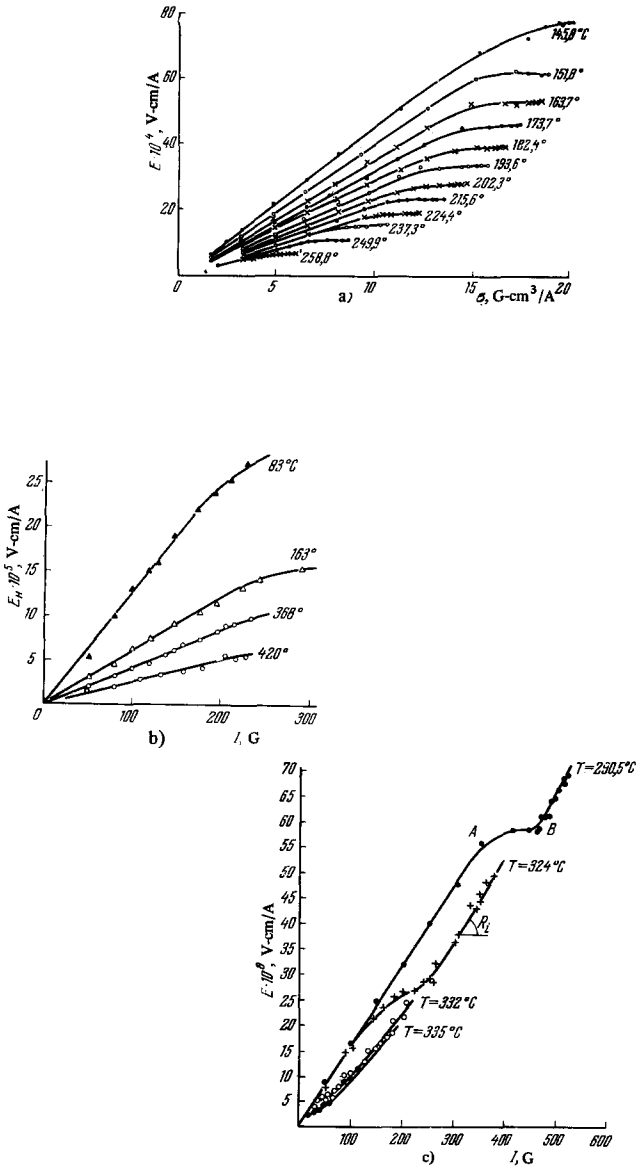

FIG. 12. Dependence of the Hall emf on magnetization for various temperatures for: a) manganese ferrite $Mn_{1.18}Fe_{1.82}O_4$, b) cobalt ferrite $Co_{0.96}Fe_{2.04}O_4$, c) alloy of Invar type: 36% Ni, 58% Fe, and 6% Cr.

In ferrites we have to do with a paraprocess of two sublattices; therefore in this case we can expect a more complicated behavior of the Hall emf during the paraprocess. Different sublattices may make contributions of different signs to the Hall emf in the paraprocess region. According to data of our measurements, the sign of the spontaneous Hall coefficient in ferrites is not determined by the sign of the current carriers. For example, in magnetite Fe_3O_4 and in nickel ferrite $NiFe_2O_4$ the basic current carriers are the electrons; yet the spontaneous Hall coefficient R_S of magnetite has a negative sign, whereas the coefficient R_S of nickel ferrite has a positive sign. As for the sign of the spontaneous Hall coefficient for metals, with respect to this question there are in the literature contradictory data[4,9,10,13,39,61,99].

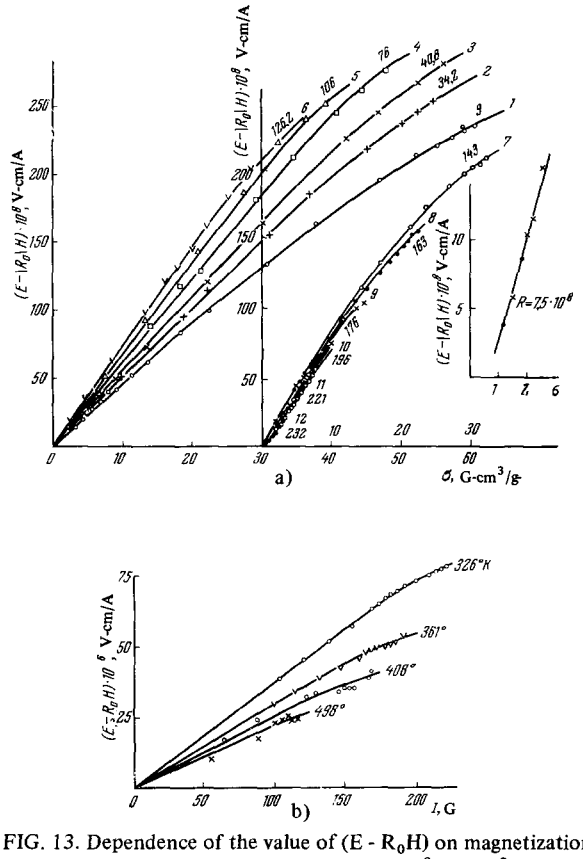

FIG. 13. Dependence of the value of (E - R_0H) on magnetization for various temperatures: a) Fe_3Pt alloy: 1 from 9° to 126°C; 2 – from 143° to 232°C; 3–for 221 and 232°C (the numbers on curves 1-12 are temperatures in °C); b) manganese ferrite ($Mn_{0.87}Fe_{2.13}O_4$).

From the few experimental results presented, it is impossible to draw unambiguous conclusions in regard to the behavior of the Hall effect in the paraprocess region, either for ferrites or for metals. This problem requires its own future experimental and theoretical investigation.

5. ON THE NATURE OF ELECTRICAL CONDUCTIVITY IN FERRITES

At present, two mechanisms of transport of electricity in oxide semiconductors are discussed: the jump and band mechanisms. According to the jump-mechanism model (the Verwey model), electrical conduction in the oxides of transition metals comes about through exchange of electrons between ions that have different valences and are located at crystallographically equivalent sites. In the presence of an electric field, the random character of the jumps of electrons from one ion to another becomes ordered, and a current arises in the direction of the field. In this case it is supposed that the number of current carriers is almost independent of temperature (for example, the concentration of Fe^{2+} ions) and that the decrease of electrical resistivity with increase of temperature is due to the exponential rise of the mobility of the current carriers.

The band description of electrical conduction in oxide semiconductors is based on ideas about the motion of collectivized electrons, whose number n increases with temperature according to the law

$$n = n_0 \exp\left\{-\frac{\Delta E_n}{kT}\right\},$$

where ΔE_n is the energy of activation of the carriers and describes the width of the forbidden band. Interaction of the current carriers with the lattice oscillations leads to a decrease of the mobility of the carriers with increase of temperature.

The existing experimental papers on investigation of the temperature dependence of the electrical properties of oxide semiconductors can be divided into three groups:

1. The first of these investigated the temperature dependence of the resistivity[68-73] of oxide compounds with attention to the different distribution of metallic cations of different valences in the crystal lattice. Since the conductivity is a function of the number of current carriers n and of their mobility μ, which depend simultaneously on temperature, it is clear that from the temperature dependence of the electrical resistivity alone it is impossible to draw an unambiguous conclusion about the nature of the mechanism of electrical conduction in the materials under study.

2. The electrical resistivity and the thermoelectromotive force were investigated simultaneously as functions of temperature. From data on the thermal emf, attempts have been made to determine the sign and number n of the carriers. For example, Morin[70,71] studied in detail the electrical resistivity and thermal emf of NiO and of α-Fe_2O_3 in the temperature range from 300° to 1100°C. He established that the fundamental current carriers in NiO are holes, and in α-Fe_2O_3 electrons; and that the mobility of the carriers, determined from data on the electrical resistivity and the thermal emf, is unusually small, of order 10^{-3} to 10^{-6} cm^2/V sec, and increases with rise of temperature.

By studying the thermal emf and electrical conductivity of nickel ferrite ($Ni_{0.8}Fe_{2.2}O_4$), Morin and Geballe[74] reached the conclusion that the temperature hysteresis of the electrical properties of this ferrite is due not to a change of the number of carriers, but to hysteresis in the mobility of the carriers, and that the conduction arises through a jump mechanism. Similar opinions are held by Lord and Parker[75] in papers on the study of the effect of oxygen pressure on the electrical conductivity of the ferrite $NiFe_2O_4$.

Heikes and Johnston[76] and Van Houten[77] thought that the increase of the electrical conductivity of NiO with rise of temperature was caused solely by the increase of mobility of the current carriers. Jonker and Van Houten[78,79], for cobalt ferrites, calculated the mobility of the current carriers on the assumption that the number of carriers is constant. The numerical values of the mobilities of the electrons and of the holes, at room temperature, were 10^{-4} and 10^{-8} cm^2/V sec respectively; the mobility increased exponentially with rise of temperature. On this basis the authors asserted that for description of the electrical properties of cobalt ferrites, the usual band theory is inapplicable, and that the temperature dependence of the mobility of the current carriers is in agreement with the concept of localized energy levels. At the same

time Sinna and Kishan Pran[80], who measured the thermal emf and electrical conductivity of the ferrite $Ni_{0.2}Cu_{0.8}Fe_{1.9}Mn_{0.1}O_4$ in the temperature range 0° to 300°C, proposed that in the ferrite studied by them, a 3d conduction band is formed, in consequence of which the current carriers have a very small mobility and a large effective mass, and that the motion of the current carriers comes about by a band mechanism of conduction.

Thus in the experimental papers belonging to the second group (study of the electrical resistivity and thermal emf of oxide semiconductors), contradictory points of view are expressed on the nature of the mechanism of conduction in these substances.

The very small values of the mobility of the current carriers and the deductions from experimental data on its increase with rise of temperature have demanded theoretical explanation. The first work in this direction was done by Yamashita and Kurosawa[81] and later by Holstein[82] (see also[83]).

Jump of an electron from an ion to an ion of different valence means, from the point of view of quantum mechanics, that the electron should be described by a wave function that is localized near one of the ions in whose neighborhood it can be (see, for example, papers of Nagaev[84], Dogonadze and Chizmadzhiev[85], Dogonadze, Chernenko, and Chizmadzhiev[86], Klinger[87], Lang and Firsov[88], and others). Allowance for the possibility of a transition of the electron from one lattice site to another causes a broadening of the levels into narrow bands. If we suppose that the current carriers are polarons (electron + lattice polarization), then in such a model the mobility of the carriers can be very small, and it should have an activation character.

Klinger[87] solved the polar problem with allowance for strong electron-phonon interaction. According to the theory of motion of a polaron of small radius, the drift mobility of the carriers may differ from the Hall mobility both in magnitude and in temperature dependence[89].

It is necessary to investigate the question whether the oxides NiO, CoO, MnO, and the ferrites are materials in which the mobility of the carriers is described by the theory of polarons of small radius (see later on this).

3. The third group of papers is devoted to the investigation of the Hall effect, optical and photo-optical phenomena, electrical resistance, and the thermal emf in the oxides NiO, CoO, MnO, and the ferrites[48,59,90-98]. These investigations first of all made it possible to establish that the Hall and drift mobilities almost coincide numerically and decrease with rise of temperature. Such experimental results contradict deductions from papers of the second group.

Investigations of the optical absorption spectra and of photoconductivity[92,39] enabled Ksendzov to determine the width of the forbidden band in NiO (4 eV) and in MnO (3 eV). The width of the conducting filled band according to Ksendzov's data is of order 1 eV. In addition, for NiO the value of the drift mobility of holes and of electrons was determined by the method of optical excitation[39,40]. The agreement of the drift mobility of holes under optical excitation with the mobility of holes excited by phonons in NiO with Li impurity also serves to confirm the electronic energy-spectrum scheme in NiO proposed in paper[92].

The agreement of the Hall and drift mobilities implies the possibility of determining the number of carriers n from the classical Hall coefficient R_0. Figure 14 shows, for CoO, the dependence of the electrical conductivity and of the concentration and mobility of the current carriers, calculated from the Hall effect, on the reciprocal of the temperature[93]. It is seen that the concentration of current carriers depends strongly on temperature and that the conduction is caused by carriers of a single sign. Estimation of the drift mobility of holes in the high-temperature region (500–1100°C) in CoO also gives a value close to the Hall value (see curves 3 and 3′ in Fig. 14).

In[94] it was also shown that in the impurity-conductivity range, the behavior of the electrical conductivity in CoO is basically determined by the concentration of the current carriers, not by their mobility. Consequently, the supposition made in papers of the second group about the weak temperature dependence of the number of carriers in NiO, CoO, etc. is groundless.

Bosman and Crevecoeur[98] and also Austin, Springthorpe, and Smith[97], on the basis of measurements of the Seebeck and Hall effects and of the electrical conductivity, reached the conclusion that for NiO (with impurity Li), application of the jump model is incorrect. Qualitatively, the results of their papers support the fundamental conclusions of Ksendzov's paper.

As regards the ferrites, the explanation of the mechanism of their electrical conduction is a still more difficult problem, since in these materials one must expect a more complicated energy-band structure than for NiO and CoO.

In[55,100-103], measurements were made of the Hall emf in magnetite and several ferrites. The concentration of the current carriers and their mobility were calculated only in the neighborhood of room temperature. The authors of the present review have conducted an investigation of the temperature dependence of the Hall emf for a number of ferrites and have obtained the temperature dependence of the concentration of the conduction electrons and their mobility. The method proposed in[43] for determining the classical Hall coefficient R_0 made it possible to eliminate the effect of the paraprocess on the results of a determination of R_0. Figure 15 shows data for Mn ferrite with various contents of hausmannite[59]. It is seen that over a wide temperature range (from room tempera-

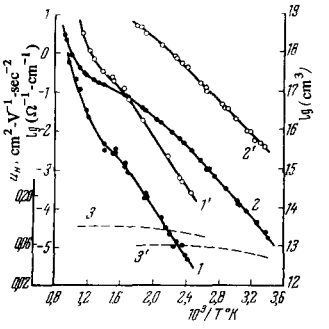

FIG. 14. Temperature dependence of the electrical conductivity (1, 1′) and of the concentration (2, 2′) and the Hall mobility (3, 3′) of holes in two specimens of cobalt oxide.

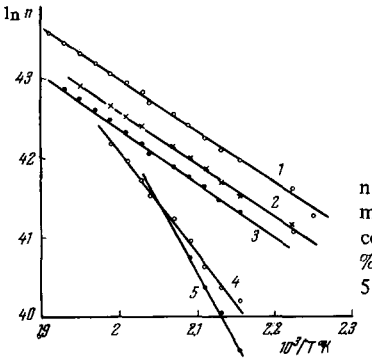

FIG. 15. Dependence of ln n on 1/T for monocrystals of manganese ferrites with various contents of Mn_3O_4. In weight %: 1, 1.36; 2, 3; 3, 8.1; 4, 15.3; 5, 22.

ture to the Curie temperature, 600°K), the carrier concentration increases exponentially with rise of temperature. With replacement of Fe ions by Mn ions, the activation energy rises and the number of current carriers decreases. The Hall mobility decreases with rise of temperature (Fig. 16). The exponential increase of number of carriers and the decrease of their Hall mobility with rise of temperature indicate that in the temperature interval considered, jumping of electrons between iron ions of different valence, in the Verveĭ manner, does not occur in the electrical conduction of ferrites. It may be supposed that the decrease of mobility with temperature is caused by interaction of electrons not only with phonons but also with magnons. In the same temperature range, the increase of iron ions in the lattice of manganese ferrite leads to some slowing down of the rate of decrease of mobility with rise of temperature. At the same time, the abundance of iron ions in the manganese ferrite raises the Curie point. A rise of the Curie point is equivalent to a diminution of the scattering on magnetic inhomogeneities of the spin system. In this connection, there is interest in measurements of the Hall effect in ferrites over a wider temperature range than has been done hitherto. Svirina and collaborators[48] have investigated the Hall and Nernst-Ettingshausen effects in monocrystalline manganese, nickel, and cobalt ferrites in the temperature interval 77°–600°K. It is seen in Fig. 17 that the temperature dependence of the Hall mobility of Mn, Ni, and Co ferrites over a wider temperature interval has a complicated form (from 77° to 150°K). It increases a little, reaches a small maximum, and then decreases with rise of temperature. It follows that in ferrites there is a complicated mechanism of scattering of carriers[63]. Apparently ferrites do not belong to the class of materials with small values of the mobility of the current carriers; that is, instead of the increase expected according to the theory[89], there is observed a decrease of the Hall mobility with increase of temperature.

The concentration of current carriers, calculated from the classical Hall coefficient R_0, depends exponentially on temperature over a wide temperature range (Fig. 18). Consequently, the suppositions of a number of authors[70,74,113] about weak dependence of the number of current carriers on temperature in ferrites (based on the data of Hall-effect measurements) are not substantiated.

It should be remarked that the temperature dependence of the thermal emf does not give a complete picture of the change of the number of carriers with temperature. For example, for monocrystalline Mn ferrites and for polycrystalline lithium ferrite the thermal emf increases slightly with rise of temperature. Yet the carrier concentration n, calculated from the Hall effect, increases meanwhile according to an exponential law (see Fig. 18). If one uses the formula for the thermoelectromotive-force coefficient α proposed by Jonker[79], and correct for the jump mechanism of conduction, and if one calculates the number of carriers by this formula, then one gets a discrepancy of two to three orders of magnitude between the values of n obtained from α and from R_0. For this reason the drift and Hall mobilities of the carriers for ferrites differ in value by about two or three orders, the Hall mobility being larger than the drift. Several authors[104,105] see a correspondence with experiment

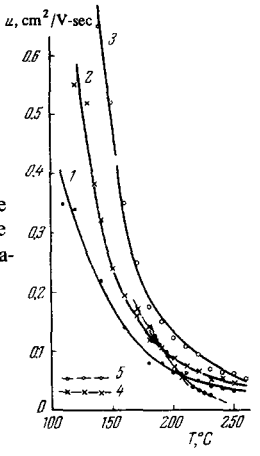

FIG. 16. Temperature dependence of the Hall mobility for monocrystals of manganese ferrites with various contents of Mn_3O_4; notation the same as in Fig. 15.

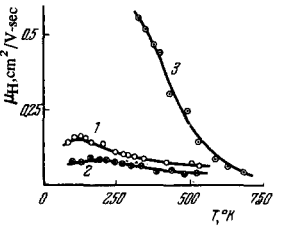

FIG. 17. Temperature dependence of the Hall mobility for manganese, nickel, and cobalt ferrites. 1–$Mn_{0.87}Fe_{2.13}O_4$; 2–$Ni_{0.69}Fe_{2.31}O_4$; 3–$Co_{0.94}Fe_{2.06}O_4$.

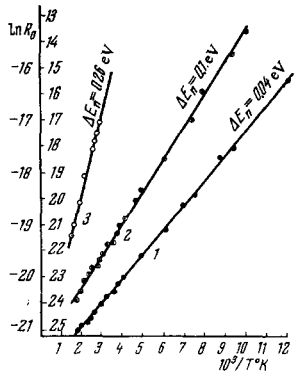

FIG. 18. Dependence of ln R_0 on 1/T for various ferrites: 1–$Mn_{0.87}Fe_{2.13}O_4$; 2–$Ni_{0.69}Fe_{2.31}O_4$; 3–$Co_{0.94}Fe_{2.06}O_4$.

in the theory of the Hall effect for materials with small mobility of the current carriers[89].

In our opinion and in the opinion of other authors (Ya. M. Ksendzov, V. P. Zhuze), the reason for the disagreement in the numerical values of the quantity n as determined from the thermal emf and from the Hall effect for ferrites is an incorrect choice of formulas for the thermal emf (correct for the jump mechanism of electrical conduction). Furthermore, the formulas for the thermal emf were derived for nonmagnetic semiconductors and therefore do not take into account the mechanism of scattering on magnons. Incidentally, the thermal emf for ferrites should have a spontaneous magnetic part, analogous to the magnetic part of the electrical resistivity.

Guverich and Nedlin[106,107] and Guverich and Korenblit[108] considered theoretically the peculiarities of the thermal emf in ferromagnetic metals. They showed that for these materials the thermal emf consists of three parts: electron, phonon, and magnon. For example, if one takes account of scattering only on defects, then, according to the results of these authors, scattering of electrons on spin waves gives a value of the thermal emf appreciably exceeding the usual thermal emf. Since there is at present no theory of the thermal emf for ferromagnetic semiconductors, attempts to obtain information about the mechanism of electrical conduction in them from data on the thermal emf cannot give positive results.

Thus, analysis of experimental data on the Hall effect in ferrites allows us at present to draw the following conclusions:

1. The temperature dependence of the spontaneous Hall coefficient in ferrites is determined by the change in the magnetic part of the electrical resistivity; that is, by the change in the concentration of the current carriers and in their scattering on magnetic inhomogeneities. This deduction follows from the linear dependence of the ratio R_S/R_0 on I_S^2 in ferrites.

2. The temperature dependence of the electrical resistivity of ferrites is determined not by the mobility of the carriers, but basically by the change in their number, since the concentration of current carriers increases exponentially with increase of temperature, whereas the Hall mobility as a rule decreases. This fact contradicts the Verwey model of electrical conduction in ferrites and implies the possibility of explaining it from the point of view of the band model.

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