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Self-reversal of the magnetization of natural and synthesized ferrimagnets

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Contents

Abstract. According to modern concepts, the reverse magnetization of rocks is a result of polarity reversals of the terrestrial magnetic field. However, rocks sometimes become magnetized antiparallel to an applied field during laboratory experimentation. This phenomenon is called self-reversal of magnetization. Self-reversal is also observed when magnetizing synthesized ferrimagnets. This poses the challenge of determining the true nature of the reverse magnetization of rocks. Discussed in this review devoted to investigations into self-reversal of rock magnetization, with emphasis on the physical mechanisms of self-reversal, are both the experimental and theoretical studies of the authors, including results obtained in terms of the model they developed based on the Néel N type self-reversal mechanism. A comparison of computer simulations with experimental results is also presented.

1. Introduction

This review is devoted to an unusual physical phenomenon — magnetization of ferrimagnetic materials in a direction antiparallel to the magnetizing field. This phenomenon is called the self-reversal of magnetization. The self-reversal is usually observed in ferrimagnetic materials (see Sections 2.3 and 3) in relatively weak magnetic fields [1, 2].

Since the magnetic field of the Earth has a small strength ($H_{\mathrm{GM}} \approx 0.5$ G [3]), the self-reversal is most frequently observed, as follows from an analysis of the literature, in ferrimagnetic minerals of rocks. The problem of the self-reversal of natural remanent magnetization (NRM) I_{n} of rocks is of global and fundamental significance for geophysics, since the NRM of rocks contains the main information that permits one to estimate the magnitude and direction of the ancient geomagnetic field (GMF) and, thereby, the evolution of the GMF, which is inseparably linked with the evolution of our magnetic planet [4].

All bodies — both macroscopic and microscopic — offer magnetic properties to some extent. In the microscopic world, the carriers of magnetism are atoms and nuclei, in which electrons and nucleons possess elementary magnetic moments. It is the interaction of these moments on a microscopic level that mainly determines the magnetic properties of macroscopic objects. On a planetary scale, all extraterrestrial objects, including the Sun, Moon, stars, and all the planets of the solar system (except for Pluto, for which no data on its own magnetic field are available at present), possess magnetic fields. Moreover, there exists an interplanetary magnetic field, and even interstellar space is characterized by a galactic magnetic field [3].

The Earth has its own magnetic field. The geomagnetic field penetrates the entire Earth, oceans and atmosphere, acting on both biotic and abiotic environments. The geomagnetic field magnetizes all rocks, especially efficiently during

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their formation, when extremely favorable conditions for a sharp increase in the magnetic susceptibility of rocks arise. The force lines and the strength of the GMF change continuously. Variations in the GMF have both long and short time periods equal to several hundred or thousand years and from several months to a few fractions of a second [5]. In addition, there exists a tendency toward a shift of the force lines of the GMF to the west at a rate of 0.2° per annum (west drift). The long-period variations (with a period of $T = 60-1800$ years or greater) are called secular; short-period variations ($T < 1$ year) are quite various in both duration and nature. According to modern concepts [3], the sources of secular variations are located in the Earth's core, and the sources of short-period variations in the upper layers of the atmosphere (ionosphere and magnetosphere). The intensity of short-period variations depends on the activity of solar – terrestrial interactions. The most intense geomagnetic signals are called storms and substorms. The solar – terrestrial interactions are also the cause of auroras.

In the magnetosphere, at altitudes from two to six Earth's radii, there exist radiation belts confining high-energy charged particles. It is quite possible that in the absence of these distinctive magnetic traps (at $H_{\mathrm{GM}} = 0$) such high-energy particles could reach the Earth's surface and exert a significant effect on its biosphere [6].

Thus, the GMF is of global ecological significance not only since it affects the biosphere and noosphere of the Earth, but also since it is a barrier to the penetration of high-energy extraterrestrial particles to the Earth's surface.

Geomagnetic investigations performed at the beginning of the twentieth century (see Section 2) revealed that rocks of different geological ages had NRM that was oriented both along the direction and opposite to the direction of the modern GMF [4]. This suggested that in past geological epochs the GMF repeatedly changed its polarity, i.e., GMF reversals occurred, during which the strength of the GMF approached zero. This opened the problem of the effect of reversals on the evolution of the GMF and the Earth in the past and in the future. However, the problem of GMF reversals has not been solved completely to date. As indicated above, there exist physical mechanisms of self-reversal (inversions) of the rock NRM itself, in particular, the so-called self-reversal of magnetization of ferrimagnetic minerals which mainly determine the magnetism of rocks.

This review is devoted to the very important above-mentioned problems that are of both purely physical interest and an undoubted global geophysical significance.

2. The principal properties of the geomagnetic field and related geophysical phenomena

2.1 The modern model and the structure of the geomagnetic field

We first consider the principal properties of the GMF near the Earth's surface. According to the Gauss general theory of geomagnetism [7], the magnetic potential of the GMF can be represented in the form of an infinite series of spherical harmonic functions:

$$U = \sum_{n=1}^{\infty} \sum_{m=0}^{n} \frac{R^{n+2}}{r^{n+1}} \left(g_n^m \cos m\lambda + h_n^m \sin m\lambda \right) P_n^m(\cos\theta) , \tag{2.1}$$

where R is the Earth's radius, r is the distance from the center of the Earth to the point of observation with geographic coordinates (θ, λ, $\theta = \pi/2 - \varphi$), $P_n^m(\cos\theta)$ are the associated Legendre functions, and g_n^m and h_n^m are the constant Gauss coefficients. The components (elements) of the geomagnetic field are determined by differentiation in corresponding directions

$$X = -\frac{\partial U}{r\,\partial\theta} \text{ (northern)} ,$$
$$Y = -\frac{\partial U}{r \sin\theta\, \partial\lambda} \text{ (eastern)} ,$$
$$Z = -\frac{\partial U}{\partial r} \text{ (vertical)} . \tag{2.2}$$

In view of the continuous changes in the GMF, the International Association of Geomagnetism and Aeronomy every five years approves a new required limited set of Gauss coefficients which to the greatest accuracy determine the modern GMF measured in geomagnetic observatories and by satellites. At present, the number of terms in the series (2.1) that describes the model of the GMF is restricted to $n = 10$, which yields 120 Gauss coefficients [5]:

$$U = \sum_{n=1}^{10} \sum_{m=0}^{n} \frac{R^{n+2}}{r^{n+1}} \left(g_n^m \cos m\lambda + h_n^m \sin m\lambda \right) P_n^m(\cos\theta) . \tag{2.3}$$

This model is called the International Geomagnetic Reference Field. The Gauss model makes it possible to analyze the structure of the geomagnetic field which typically consists of the main geomagnetic field (dipole plus nondipole parts), an anomalous field produced by magnetized rocks, and an external magnetic field.

However, Eqn (2.3) describes only the main geomagnetic field and enables us to separate in it only the dipole part ($n = 1, m = 0, 1$) and nondipole part (at n from 2 to 10, and m from 0 to n). The anomalous field comprises only about 3% of the GMF, and the external field far less than 1%.

The dipole part comprises more than 90% of the entire GMF; therefore, to the first approximation the GMF is regarded as the field of the central dipole inclined to the rotation axis of the Earth by 10° – 12°. The anomalous part of the GMF can easily be determined by subtracting the main GMF defined by Eqn (2.3) from the field measured at a given point with coordinates (φ, λ).

2.2 Origin of the geomagnetic field

The equations of magnetic hydrodynamics and the idea of a dynamo effect, which was first suggested in Ref. [8], lie at the basis of the modern theory of the generation of the main GMF. It is assumed that the generation of the GMF from the original very weak magnetic field, as well as the generation of all secular variations, can arise due to motions in the conducting liquid core of the Earth, in particular, due to the lag of the upper layers of the core from the Earth's mantle upon the Earth's rotation, which is also confirmed by the west drift of the GMF [5].

The Maxwell equations make it possible to write down an equation for the induction of the magnetic field **B** in the following form

$$\frac{\partial \mathbf{B}}{\partial t} = (\mu\sigma)^{-1} \nabla^2 \mathbf{B} + \mathrm{rot}\,[\mathbf{v} \times \mathbf{B}] , \tag{2.4}$$

where $(\mu\sigma)^{-1}$ is the magnetic viscosity. The velocity $\mathbf{v}$ is determined from the Navier–Stokes equation of motion of a unit liquid volume:

$$\frac{\mathrm{d}\mathbf{v}}{\mathrm{d}t} = -\frac{1}{\rho}\,\mathrm{grad}\,p + \mathbf{g} + \nu\nabla^2\mathbf{v} - 2[\boldsymbol{\omega}\times\mathbf{v}] + \frac{1}{\mu\rho}\,[\mathrm{rot}\,\mathbf{B}\times\mathbf{B}]\,, \tag{2.5}$$

where

$$\frac{\mathrm{d}\mathbf{v}}{\mathrm{d}t} = \frac{\partial\mathbf{v}}{\partial t} + \mathbf{v}\,\mathrm{grad}\,\mathbf{v}\,,$$

ρ is the density, p is the pressure, $\mathbf{g}$ is the acceleration of gravity, ν is the kinematic viscosity, $2[\boldsymbol{\omega}\times\mathbf{v}]$ is the Coriolis force, and $1/(\mu\rho)[\mathrm{rot}\,\mathbf{B}\times\mathbf{B}]$ is the force of electromagnetic interaction.

To equations (2.4) and (2.5), we should add a continuity equation

$$\frac{\partial\rho}{\partial t} + \mathrm{div}\,(\rho\mathbf{v}) = 0\,, \tag{2.6}$$

which at $\rho = \mathrm{const}$ reduces to

$$\mathrm{div}\,\mathbf{v} = 0\,. \tag{2.7}$$

The simplest initial approach to the problem of the generation of the main GMF consists in constructing kinematic models of dynamos in which the velocity of liquid is assumed to be given, and only $\mathbf{B}$ is determined from Eqn (2.5). The first dynamo models appeared in 1958 [6]. Models of a uniform dynamo [6], a turbulent dynamo [6], etc. have been developed. The heart of the problem is in finding a scheme of transformation and enhancement of the originally very weak magnetic field to the level of the modern poloidal magnetic field observed on the Earth's surface. To this end, the so-called ω effect and α effect are used in the theories of the kinematic dynamo. The ω effect is responsible for the generation of a sufficiently strong (100–300 G in the Earth's core) toroidal field from the originally very weak poloidal field due to the rotation of layers of the liquid core relative to the Earth's mantle. Then, as a result of the α effect, a poloidal field (30–40 G in the Earth's core, and 0.5 G on the Earth's surface) is generated from the above toroidal field. The generation of the poloidal field from the toroidal one occurs as a result of convection in the core. The dynamics of the core are determined by Lorentz and Coriolis forces. The dynamo models that utilize both effects are called $\alpha\omega$-dynamo models [9]. In principle, a toroidal field can also arise from a poloidal field according to the α mechanism. Such dynamos are called α^2 dynamos [9].

The models of a hydromagnetic dynamo used at present are kinematic. The dynamics and the energy-related mechanisms in such models remain absolutely unknown. Therefore, no theory on the origin of the magnetic field of the Earth that could satisfactorily describe the observed features of the GMF exists at present.

A more natural and plausible mechanism for the generation of the GMF is that caused by toroidal currents flowing in the Earth's core. If we take into account that the magnetic dipole of the Earth is oriented from north to south, the toroidal current must be directed west, i.e., precisely in the direction of the west drift of the GMF. This point of view is taken to be more reasonable by one of the authors of this review.

2.3 The paleomagnetic model of the geomagnetic field

Measurements of the GMF have been performed only for the last 400 years. The data obtained are obviously insufficient for studying secular variations with time periods of 600, 900, and 1800 years, or greater, and, moreover, for understanding the evolution of the GMF which has existed about four billion years. Therefore, a so-called paleomagnetic method (PM) of studying the ancient GMF was developed in the mid-1950s, which is based on measurements of the natural remanent magnetization of igneous and sedimentary rocks of corresponding geological ages [4].

The magnetization $\mathbf{I}$ of rocks consists of two components [10]:

$$\mathbf{I} = \varkappa_0\mathbf{H}_{\mathrm{GM}} + \mathbf{I}_{\mathrm{NRM}}\,, \tag{2.8}$$

where $\varkappa_0$ is the initial magnetic susceptibility of the ferrimagnetic fraction of the rock, I_{NRM} (or I_{n}) is the natural remanent magnetization measured at $H = 0$, and $\varkappa_0\mathbf{H}_{\mathrm{GM}}$ is the reversible magnetization induced by the GMF, which is equal to zero at $H_{\mathrm{GM}} = 0$.

The natural remanent magnetization of igneous rocks is formed during the cooling of ferrimagnetic minerals in the GMF after their crystallization from magma to the temperature of the Earth's surface. The process of magnetization of a ferrimagnet (by definition, a ferrimagnet constitutes a substance involving two or more uncompensated magnetic sublattices) that occurs during its cooling from $T = T_{\mathrm{C}}$ (T_{C} is the Curie point) to the temperature of the Earth's surface is called thermo magnetization; the magnetization I_{T} acquired as a result of this process is called thermomagnetization (TM), and that obtained at $H = 0$ the thermoremanent magnetization (TRM) I_{rT}. This process can easily be reproduced under laboratory conditions. During thermomagnetization in a field H there arise TM and TRM whose magnitudes far exceed (by an order of magnitude or even greater) the isothermal remanent magnetization (IRM) formed upon magnetization in a field H at $T = \mathrm{const}$.

The natural remanent magnetization can also emerge as a result of chemical transformations [chemical remanent magnetization (CRM)], the action of enhanced pressures (piezoremanent magnetization), and other processes, but the most common NRM of igneous rocks is associated with the TRM [11]. With time, the erupted rocks become broken and disintegrate (down to nanoparticles). The rock particles can be transferred by water flows to various basins (lakes, seas). In the basin, the fine magnetized ferrimagnetic particles become oriented along the GMF direction while falling to the bottom. Mixing with particles of other compositions, they form sediments and, later, sedimentary rocks. Thus, the so-called orientational (depositional) magnetization of sedimentary rocks arises, which, just as the TRM of erupted rocks, contains paleomagnetic information.

Based on the paleomagnetic data, it is obviously impossible to describe the ancient GMF to the same degree of accuracy to which we can describe the modern GMF. Therefore, the simplest model of the GMF is applied in paleomagnetic investigations, i.e., the field of the central dipole directed along the Earth's rotation axis. In this model, a single Gauss coefficient [5]

$$g_1^0 = \frac{M}{R^3}\,, \tag{2.9}$$

is utilized, where M is the magnetic moment of the Earth's dipole. In the paleomagnetic method, it is assumed that the NRM vector of the rock of a certain geological age reflects on the average the direction of the vector of the ancient GMF in which the rock has been magnetizing. With this assumption, we can determine the declination D_{anc} and inclination J_{anc} of the NRM and, thereby, of the ancient GMF by measuring the components of the NRM vector in a sufficiently large (to ensure good statistics) number of samples (Fig. 1a).

In the paleomagnetic model, the elements of the GMF are written down as

$$X = \frac{M}{R^3}\cos\varphi_{\text{anc}}, \quad Y = 0\,, \quad Z = \frac{2M}{R^3}\sin\varphi_{\text{anc}}\,, \tag{2.10}$$

where φ_{anc} is the ancient (corresponding to the instant of action of the ancient GMF) geographic latitude of the site of sampling. The following formula for determining φ_{anc} can be obtained from Eqn (2.10):

$$\tan J_{\text{anc}} = \frac{Z}{X} = 2\tan\varphi_{\text{anc}}\,, \quad \varphi_{\text{anc}} = \arctan(0.5\tan J_{\text{anc}})\,. \tag{2.11}$$

The ancient latitude φ_{anc} does not coincide frequently with the modern latitude φ of the site of sampling (Fig. 1b), which can be related to both the change in the position of the paleomagnetic pole and to a tectonic displacement of the block of the Earth's crust in which the sampling site is located. Indeed, the migration curves of the paleomagnetic pole, obtained in many works, proved to be various for different continents (Fig. 2). This is the first fundamental result of paleomagnetic investigations.

However, in the model of the paleomagnetic field there exists a rigid tie between the paleomagnetic and geographic poles. Therefore, based on the data on the motion of the paleomagnetic pole we can arrive at a conclusion that the position of the geographic pole also changed substantially (by more than 90°) in the last 400 – 600 million years. Modern astronomical and geophysical data allow no such a large deviation in the geographic pole. Therefore, the current interpretation of the curves of migration of the paleomagnetic poles is based on the assumption that the geographic and geomagnetic poles remained immobile, while related blocks of the Earth's crust (continents, lithosphere plates, etc.) were moving. The latter assumption explains the noncoincidence of the modern and ancient latitudes of the sampling sites of paleomagnetic samples [5].

The second fundamental result of paleomagnetic investigations is that in all types of ancient rocks with an age of 400 – 600 million years there was revealed an alternation of rock layers with reverse and direct (with respect to the orientation of the modern geomagnetic field) NRMs.

The reverse (negative) magnetization in the northern hemisphere is assumed to be the one that is directed into the

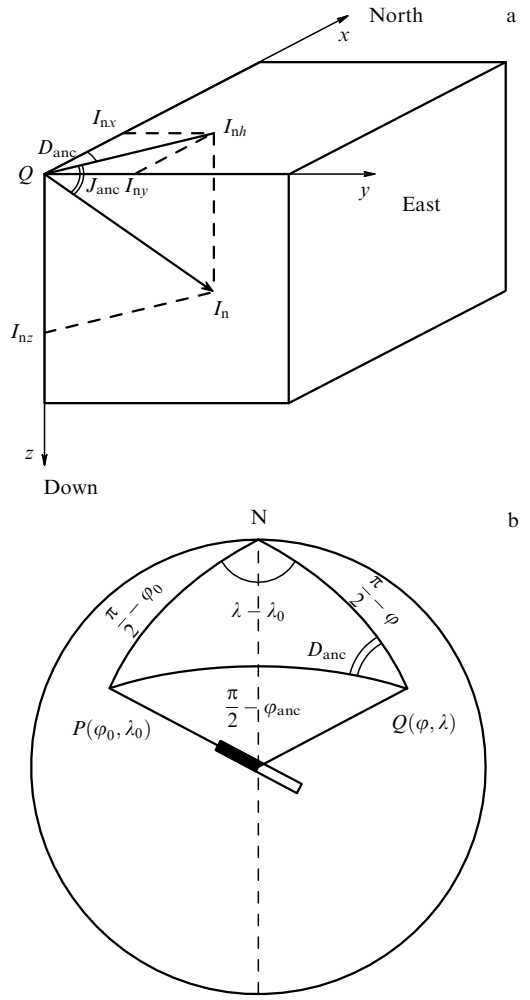

Figure 1. (a) Components of the natural remanent magnetization (NRM) of a rock at some point of the Earth's surface $Q(\varphi, \lambda)$, where φ is the latitude. and λ is the longitude. The vector I_n of the NRM shows the direction of force lines of the ancient geomagnetic field at the point Q. The angles D_{anc} and J_{anc} correspond to the declination and inclination of the ancient GMF. (b) Model of a central axisymmetric geomagnetic dipole that is employed in the paleomagnetic method: N is the northern geographic pole, $Q(\varphi, \lambda)$ is the site at which the sample for paleomagnetic investigations is taken, φ is the latitude, and λ is the longitude. The point $P(\varphi_0, \lambda_0)$ is the paleomagnetic pole, and φ_0 and λ_0 are its modern geographic coordinates. D_{anc} is the declination of the ancient GMF as determined from the direction of NRM in rocks.

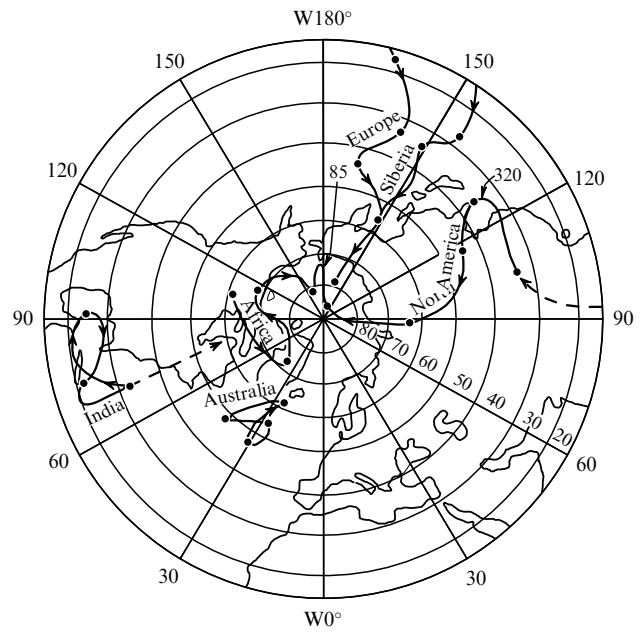

Figure 2. Migration curves of the paleomagnetic poles, obtained on the basis of measurements of the NRM of rocks from various continents.

upper (from the Earth's surface) half-space; in the southern hemisphere, it is the one that is directed into the lower half-space.

A statistical treatment of data on the directions of the NRM vectors is performed in paleomagnetic investigations, and the modern coordinates of the virtual geomagnetic pole (VGP) (see Fig. 2) are calculated by the formulas [3]

$$\sin\varphi_0 = \sin\varphi\sin\varphi_{\text{anc}} + \cos\varphi\cos\varphi_{\text{anc}}\cos D_{\text{anc}},$$
$$\sin(\lambda-\lambda_0) = \frac{\sin D_{\text{anc}}}{\cos\varphi_0}\cos\varphi_{\text{anc}}, \qquad (2.12)$$

where φ_0 and λ_0 are the modern latitude and longitude of the VGP, φ_{anc} is the ancient latitude [see Eqn (2.11)], and D_{anc} is the ancient declination corresponding to the declination of the ancient NRM vector.

The calculated [using Eqns (2.12)] coordinates of the ancient same-aged VGPs obtained by various researchers are processed statistically to obtain the modern coordinates of the VGP of a certain geological age. As a rule, the position of the reversed VGP differs by 180° (to an accuracy of a few degrees) from the position of the direct VGP of a same age. Thus, the reversals of the GMF occur along the geomagnetic axis which changes its position.

As a rule, in the investigated part of a layer of reversely magnetized rocks virtually all the samples had reverse magnetization. The layers of rocks with a direct and reverse magnetization on various continents were alternated along the vertical directed toward the Earth's surface. In the rocks of the ocean floor, the alternation of the directly and reversely magnetized rocks occurred in horizontal directions, on both sides of the mid-ocean ridges. When studying paleomagnetic samples of igneous and sedimentary rocks of a same-aged interval, taken at various continents and at various sites of the ocean floor, a similar alternation of layers with a direct and reverse NRM was revealed. For example, the equal number of changes in the sign of the NRM in same-aged lavas from California and pelagic sediments of the Pacific Ocean is observed. In many cases, a similar direction of the NRM in the sites of contact of lavas and sedimentary rocks was fixed. All this made it possible to soundly suppose that the reverse magnetization of rocks was formed in the GMF of the opposite (with respect to the modern direction) polarity, i.e., at least in the last 400 – 600 million years, reversals of the GMF (changes in its polarity) have occurred from time to time in this period, their number has been more than a thousand. The duration of such reversals is from 2000 to 6000 years. Based on the dating of the age of such reversals using radioactive-tracer techniques, a worldwide magneto-chronological scale of reversals was compiled for the continents and oceans (Fig. 3). Notice that in the case of a displacement of the paleomagnetic pole by 180° upon reversal of the GMF, the question of the rigid tie between the paleomagnetic pole and the geographic pole, which arises when the problem of a displacement of the paleomagnetic pole by 90° is discussed, is not considered by the paleomagnetists. Thus, when interpreting reversals, it is assumed that the paleomagnetic poles are displaced by 180° with respect to the immobile geographic poles, which contradicts the paleomagnetic model [5].

The authors of this review, like the majority of geophysicists, have no objection to the paleomagnetic interpretation of changes in the directions of the NRM of rocks as being due to reversals of the GMF. Note, however, that there exists

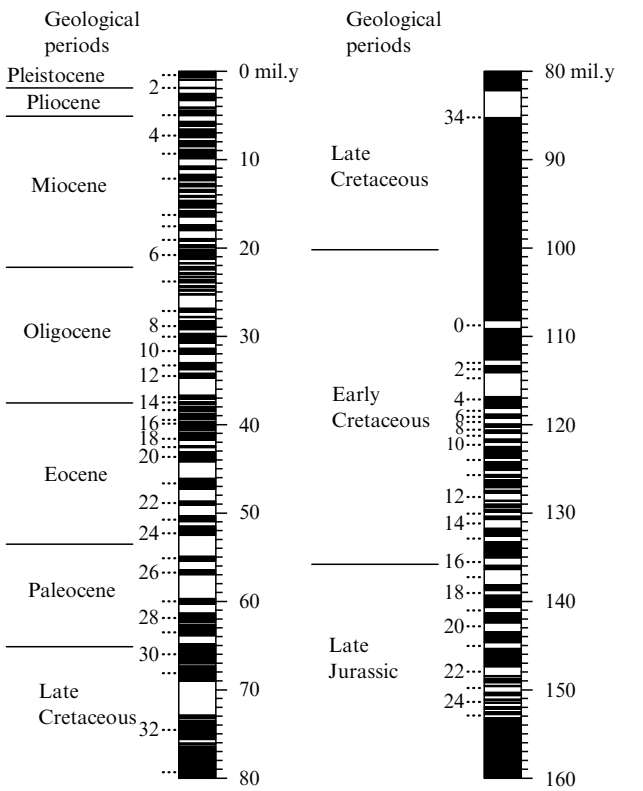

Figure 3. Magneto chronological scale of GMF used for the determination of the age of linear oceanic anomalies. To the left of the scale, order numbers of the anomalies are given; to the right of the scale, the age in millions of years. The dark regions of the scale correspond to the direct magnetization; the bright regions, to the reverse magnetization; mil.y stands for millions of years.

another mechanism for the formation of magnetization that is antiparallel to the magnetizing field, which results from some specific properties of ferrimagnets which, as is known, are the minerals that determine rock magnetism.

2.4 Fundamentals of the concept of neomobilism

The results of paleomagnetic investigations led to a host of new ideas in geophysics and geology, which are based on the concept of neomobilism. In geophysics, there have long existed two fundamentally different approaches to the consideration of the evolution of our planet, namely, fixism (stable development) and mobilism [5]. According to the mobilistic concept of the tectonics of lithospheric plates, all the Earth's lithosphere consists of 10 – 12 coarse plates which move with respect to one another with a velocity of 1 – 10 cm per year. Upon a large-scale separation (moving away) of lithospheric plates, there emerge oceans; upon a large-scale contraction, mountain ranges (massifs) appear. Mobilism, as an antithesis to fixism, sprang up in 1915 after the publication of *Die Entstehung der Kontinente und Ozeane* (*The Origin of Continents and Oceans*) by Alfred Wegener, a Deutch meteorologist, geophysicist, and polar explorer [12]. Based on the similarity of the outlines of the opposite coasts of the Atlantic Ocean and resorting to other, less known at that time, geophysical data (a hypsometric map of the Earth, the theory of isostasy, data on the velocities of seismic waves under continents and oceans), Wegener supposed that the modern structure of the continents is a consequence of the splitting up of an ancient supercontinent called Pangea into

several parts ('plates') and the separation of these parts under the effect of forces of the Earth's rotation. Between the separated fragments of Pangea (modern continents), the Atlantic, Indian, and other oceans have formed.

The fundamental ideas of the concept of plate tectonics as applied to the ocean floor were formulated in Refs [13, 14]. The essence of the ideas is that as a result of magma eruption on both sides of the mid-ocean ridges there are formed new regions of an ocean crust which symmetrically spread out from the ridges. It is assumed that such spreading is caused by the thermal convection effect in the mantle. A substantial supplement to these ideas became a hypothesis [15], formulated in 1963, which related the formation of new rocks in the centers of mid-ocean ridges (and their motion away from the ridges) to reversals of the GMF. According to the proposed hypothesis, the presence of symmetric spectra of linear sign-alternating magnetic anomalies on both sides of the mid-ocean ridges is a consequence of the expansion (spreading) of the ocean floor and changes in the polarity of the GMF.

By no means do all geologists and geophysicists support the concept of plate tectonics. According to nonadherents of the concept, there exist a number of problems that cannot be solved proceeding from the theory of plate tectonics. One prominent geophysicist of the 20th century, V A Magnitskii, wrote as long ago as 1960: "It can be said for sure that the Earth's crust separated from the shell in the process of the Earth's evolution; therefore, the experimentally established equality of heat fluxes on continents and oceans suggests that there did not exist large-scale horizontal displacements (on the order of thousands of kilometers) of large blocks of the Earth's crust supposed by the hypothesis for mobilism ... . Consequently, the continents and oceans emerged on the whole at those sites where they are located at present."

As follows from the above reasoning, virtually all the main concepts of modern geophysics, namely, the concept of plate tectonics, GMF reversal, and many other phenomena, are directly or indirectly related to or even based on paleomagnetic investigations and, first and foremost, on the magnetochronological scale of GMF reversals. Therefore, a correct interpretation of paleomagnetic data has become of progressively greater importance. Apart from the above complexities related to the interpretation of paleomagnetic poles and reverse NRM, there also exist problems related to other specific features of the paleomagnetic method [5]. First, what is measured by this method is the NRM of ancient rocks rather than the ancient GMF itself. Second, ferrimagnetic minerals are by no means always magnetized exactly along the direction of the GMF because of their anisotropy and the dependence of the internal energy of ferrimagnets on external thermodynamic conditions. There also exists the process of so-called 'magnetic aging', i.e., changes in the magnetic properties of ferrimagnetic minerals with time. The ferrimagnetic minerals of rocks evolve with time: they are oxidized, decompose, and suffer other chemical changes and transformations [10, 11]. As a result of all these natural processes, various secondary types of magnetization, such as viscous, chemical, piezo, and others, can be added to the primary NRM acquired by the rock during its formation. Consequently, substantial errors in paleomagnetic measurements can also ensure from the long residence of the initially magnetized rocks (for many thousands or millions of years) under the effect of the GMF and varying thermodynamic conditions.

Finally, as was noted in the Introduction, some part of rocks with a reverse magnetization can be formed due to spontaneous changes in the NRM polarity of ferrimagnetic minerals, i.e., as a result of the processes of self-reversal of magnetization.

Thus, there exist two opportunities for the formation of a reverse NRM: (i) as a result of GMF reversals, and (ii) as a result of the processes of self-reversal of magnetization. We will never be able to examine experimentally GMF reversals, since their duration is several thousands of years. However, by studying processes of self-reversal of the NRM of ferrimagnetic minerals and physical mechanisms of these processes, we can estimate the probability of occurrence of these processes in nature and, thereby, the probability of existence and the number of GMF reversals. It is for this reason that we present in Section 3 a review of the literature concerning the problems of self-reversal of magnetization.

3. Self-reversal of magnetization of ferrimagnets

The natural remanent magnetization whose direction is approximately antiparallel to the direction of the modern GMF (which will be further called 'reverse NRM') was for the first time discovered in igneous rocks at the beginning of the twentieth century [16, 17]. Since then, reverse NRM has repeatedly been observed in various erupted and sedimentary rocks. The existence of reverse NRM in nature has made it possible to suppose that in past geological epochs the GMF repeatedly changed its polarity. According to modern concepts, the reverse NRM of rocks is considered as having been formed in a GMF with a polarity opposite to the modern-day one.

The first rough chronological scale of GMF reversals was suggested in 1929 by M Matuyama [18] based on his investigation of more than 100 lava flows in Japan and Manchuria (a historic region in P.R. China). Later, this scale was repeatedly refined based on paleomagnetic data obtained from various continents. However, at that time the hypothesis of reversals had not yet been commonly accepted.

It was J W Graham [19] who first proposed in 1949 that rocks could acquire a reverse NRM via some physical or chemical mechanisms being alternatives with respect to GMF reversals. Graham studied a layer of sedimentary rocks of a same age to find regions with both direct and reverse NRMs. The same age of all the deposits studied permitted him to assume that the reverse NRM could not be produced in a reversed GMF, i.e., as a result of a GMF reversal. Otherwise, the entire layer would be magnetized antiparallel to the direction of the modern GMF.

Graham wrote a letter to the well-known theoretician L Néel at Grenoble and asked him whether from the theoretical viewpoint there can exist processes leading to the magnetization of magnetic materials in rocks in a direction antiparallel to the existing magnetizing field. Up to 1951, the question of the existence of physical or chemical mechanisms of acquisition of reverse NRM by rocks as an alternative to GMF reversals remained open. In 1951, Néel published his famous work [20] in which he pioneered in suggesting four physical mechanisms of acquisition of a reverse NRM by rocks. This work is fundamental for all who study self-reversal of magnetization in rocks: the conclusions of this work have served as the grounds for many researchers up to the present. Later on, this work was supplemented by the Néel publication [21].

3.1 The physics of self-reversal of magnetization processes

First, we briefly outline the physical essence of self-reversal. The energy of the magnetic moment **M** in an external magnetic field **H** is written as

$$E_H = -\mathbf{MH} = -MH\cos\varphi\,, \tag{3.1}$$

where φ is the angle between the vectors **M** and **H**. The energy E_H is smallest at $\varphi = 0$; therefore, in the field **H** the moment **M** is oriented along **H**.

However, the cases have been revealed during thermomagnetization of some natural ferrimagnets where the total thermomagnetization $\mathbf{I}_\mathrm{T}$ (and the thermoremanent magnetization $\mathbf{I}_\mathrm{rT}$) were oriented antiparallel to the field **H**. The self-reversal effect is resulted from some specific properties of the ferrimagnet itself.

When the magnetization is oriented along the field [see Eqn (3.1)], there arises a thermodynamically equilibrium magnetic state of the ferrimagnetic sample. Correspondingly, the formation in the sample of a reverse magnetization antiparallel to the field **H** should be considered as the appearance of a nonequilibrium magnetic state ('frozen' reverse TRM).

Because of the complex structure of the ferrimagnet, this state can be reached in the process of its thermomagnetization [22]. The spontaneous magnetic moment $\mathbf{M}_\mathrm{S}$ of a two-sublattice ferrimagnet in accordance with its nature initially consists of two antiparallel nonequal moments $\mathbf{M}_\mathrm{SA}$ and $\mathbf{M}_\mathrm{SB}$ of the magnetic sublattices A and B:

$$\mathbf{M}_\mathrm{S} = \mathbf{M}_\mathrm{SA} + \mathbf{M}_\mathrm{SB}\,. \tag{3.2}$$

Therefore, if $M_\mathrm{SA} > M_\mathrm{SB}$ and the total magnetization $\mathbf{M}_\mathrm{S}$ is oriented along the direction **H**, a significant part of atomic magnetic moments (see Fig. 4a) whose sum is $\mathbf{M}_\mathrm{SB}$ will be oriented antiparallel to **H**. Consider the scheme of the process of thermomagnetization, outlined in Fig. 4b. The orientation of magnetic domains in the direction of **H**, which arises at a temperature close to T_C of the ferrimagnet, is pinned by the applied magnetic field H and by the potential relief of the ferrimagnet (arising during cooling); this relief generates a spectrum of local minima of potential energy, in which appropriately oriented domains are located. At room temperature T_0 there emerges a magnetization I_T whose magnitude is determined by the degree of orientation of the magnetic moments of the domains along the direction of **H** (Fig. 4b, curve *1*). When all magnetic moments of the domains are oriented along the saturation magnetic field H_S, we have $I_\mathrm{T} = I_\mathrm{S}$. When the field H is switched off, there remains a thermoremanent magnetization. The domains that form it are held in sufficiently deep potential wells. On the average, the TRM stability can be characterized by the coercive force H_C.

The orientations and magnitudes of the magnetic moments M_SA and M_SB in the ferrimagnetic sublattices A and B are determined by the magnitudes of exchange interactions inside the sublattices (energies E_AA and E_BB) and between the sublattices (E_AB). It is essential that all these quantities (M_SA, M_SB, E_AA, E_BB, and E_AB) can vary with temperature, and the changes can be so strong that at some compensation temperature T_comp there arises such a state in the domains of the ferrimagnet that $M_\mathrm{SA} = M_\mathrm{SB}$ and $M_\mathrm{S} = 0$. At temperatures above and below T_comp, the total moment of each domain M_S will have a different orientation, since

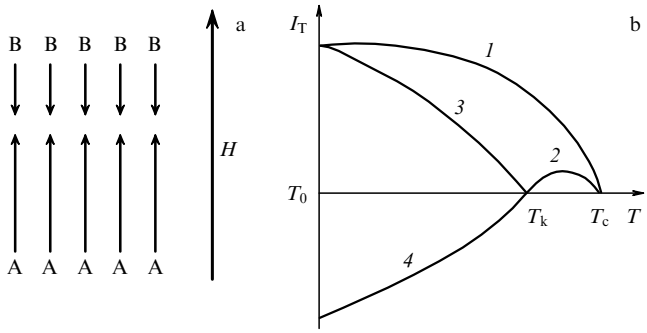

Figure 4. (a) Schematic of a location of atomic magnetic moments in A and B sublattices of a ferrimagnet. (b) Schematic of the process of thermomagnetization: (*1*) $I_\mathrm{T}(T)$ dependence for a ferrimagnet without a compensation point T_comp in the temperature range of (T_C, T_0); (*2*) $I_\mathrm{T}(T)$ dependence for a ferrimagnet with a compensation point T_comp lying in the temperature range of (T_C, T_comp); (*3*) $I_\mathrm{T}(T)$ dependence for a ferrimagnet with a compensation point T_comp lying in the temperature range of (T_comp, T_0) for $H > H_\mathrm{C}$, where H_C is the coercive force, and (*4*) $I_\mathrm{T}(T)$ dependence for a ferrimagnet with a compensation point T_comp in the temperature range of (T_comp, T_0) for $H < H_\mathrm{C}$.

$M_\mathrm{SB} < M_\mathrm{SA}$ for $T > T_\mathrm{comp}$, while $M_\mathrm{SB} > M_\mathrm{SA}$ for $T < T_\mathrm{comp}$. This also refers to the spontaneous magnetization I_S of the entire ferrimagnetic sample. The possibility of the existence of ferrimagnets in which $I_\mathrm{S}(T_\mathrm{comp}) = 0$ at a certain temperature $T = T_\mathrm{comp}$ (N type curves according to Néel's classification) was established theoretically in Ref. [22]. This work will be considered in more detail below in this section.

During the thermomagnetization of such a ferrimagnet in a temperature range $T_\mathrm{comp} < T < T_\mathrm{C}$ there emerges a thermomagnetization I_T oriented along **H**, which will first increase upon cooling from T_C and then, with approaching T_comp, decrease in view of the decrease in the I_S of the ferrimagnet; finally, it vanishes ($I_\mathrm{T} = 0$) at $T = T_\mathrm{comp}$ (Fig. 4b, curve *2*). As the cooling continues for $T < T_\mathrm{comp}$, I_T can behave in two different ways which are determined by the magnitude of I_S of the domains and by the relationship between the coercive force H_C and the external magnetic field H.

If for $T < T_\mathrm{comp}$ we have $H > H_\mathrm{C}$, the magnetization will again increase toward H during further cooling under given conditions (Fig. 4b, curve *3*), since the magnetic energy $I_\mathrm{S}HV$ (where V is the volume of the domain) will be sufficient for aligning the domains along the field direction. When $H < H_\mathrm{C}$, the domain turn is impossible because of the smallness of the energy $I_\mathrm{S}HV$, and I_T will increase for $T < T_\mathrm{comp}$ in the direction that is antiparallel to **H** due to an increase in I_S which is oriented along I_SB (Fig. 4b, curve *4*).

The process considered above (Fig. 4b, curves *2–4*) is a process of self-reversal of thermomagnetization in ferrimagnets with $I_\mathrm{S}(T)$ curves of the N type according to Néel's classification [22]. The resulting thermoremanent magnetization will be generated in a nonequilibrium state and pinned by the local energy barriers of the ferrimagnet.

An analogous change in the direction of I_S by 180° in domains may also occur due to diffusion of magnetic ions between the sublattices. Therefore, the self-reversal of magnetization is also possible as the result of chemical processes in ferrimagnetic minerals [23].

Now, we shall dwell briefly on Néel's theory of ferrimagnetism [22] and the N type curves of the temperature dependence of spontaneous magnetization, which were

mentioned above and are obtained on the basis of this theory. It should be emphasized that in 1970 Néel was awarded the Nobel Prize in Physics [24] "for fundamental work and discoveries concerning antiferromagnetism and ferrimagnetism which have led to important applications in solid state physics". In 1948, Néel developed a theory of collinear ferrimagnetism [22], in which he for the first time introduced the idea of ferrimagnetism as one type of magnetic ordering existing in nature, and of a ferrimagnet as a substance possessing two uncompensated magnetic sublattices. According to this theory, by analogy with the Weiss theory of the molecular (average) field for a ferromagnet [25], any interaction of a magnetic ion residing in the sublattice A of a ferrimagnet with nearest neighbors from the sublattices A and B can be replaced by the action of a certain 'average' field on this ion; the same refers to any magnetic ion from the sublattice B.

The total molecular fields acting on the atoms of the sublattices A and B are written down as

$$\mathbf{H}_{\mathrm{MA}} = n\alpha\,\mathbf{I}_{\mathrm{SA}} - n\mathbf{I}_{\mathrm{SB}}\,,$$
$$\mathbf{H}_{\mathrm{MB}} = n\beta\,\mathbf{I}_{\mathrm{SB}} - n\mathbf{I}_{\mathrm{SA}}\,, \tag{3.3}$$

where n, $n\alpha$, and $n\beta$ are the molecular-field constants that determine the interaction of magnetic moments inside the sublattices and between the sublattices (in what follows, we will use molecular-field constants expressed in units of n, i.e., we will denote them α and β).

Following the paper [22], we consider the crystal lattice of a ferrimagnet consisting of two nonequivalent magnetic sublattices A and B. The temperature dependences of the spontaneous magnetizations I_{SA} and I_{SB} of the sublattices A and B, respectively, are obtained by solving the following set of equations

$$\mathbf{I}_{\mathrm{SA}} = \lambda I_{\mathrm{S0}} B_J\left(\frac{Jg\mu_{\mathrm{B}} n(\alpha\,\mathbf{I}_{\mathrm{SA}} - \mathbf{I}_{\mathrm{SB}})}{kT}\right),$$
$$\mathbf{I}_{\mathrm{SB}} = \mu I_{\mathrm{S0}} B_J\left(\frac{Jg\mu_{\mathrm{B}} n(\beta\,\mathbf{I}_{\mathrm{SB}} - \mathbf{I}_{\mathrm{SA}})}{kT}\right), \tag{3.4}$$

where λ and μ are the fractions of the magnetic ions in the sublattices A and B, respectively ($\lambda + \mu = 1$); λI_{S0} and μI_{S0} are the spontaneous magnetizations of the sublattices A and B at $T = 0$ K, and B_J is the Brillouin function

$$B_J(x) = \frac{2J+1}{2J}\coth\left(\frac{(2J+1)\,x}{2J}\right) - \frac{1}{2J}\coth\frac{x}{2J}\,,$$

where J is the internal quantum number of the total moment, g is the Landé factor, μ_{B} is the Bohr magneton, and k is the Boltzmann constant.

Each of the equations (3.4) is an equation of state of a ferrimagnet in the molecular-field theory, with the argument of the Brillouin function representing the ratio of the magnetic energy to the thermal energy.

The total spontaneous magnetization $I_{\mathrm{S}}(T)$ is the vector sum of spontaneous magnetizations of individual sublattices:

$$\mathbf{I}_{\mathrm{S}}(T) = \mathbf{I}_{\mathrm{SA}}(T) + \mathbf{I}_{\mathrm{SB}}(T)\,. \tag{3.5}$$

By varying α and β in Eqns (3.4) at fixed values of λ and μ (let them be $\lambda = 1/3$ and $\mu = 2/3$) and other parameters, Néel obtained six different temperature dependences of spontaneous magnetizations (Fig. 5). Two of these (Q and R) correspond to analogous dependences of a ferromagnet; two other types of curves (M and P) are characterized by a slight decrease in I_{S} with decreasing T. The temperature dependence of I_{S} of a ferrimagnet leading to the appearance of a compensation point T_{comp}, at which $I_{\mathrm{S}} = 0$, is called, according to Néel, an N type dependence (Fig. 5b). Therefore, the physical mechanism of self-reversal related to such a dependence can be called the Néel N type mechanism. It should be emphasized that the $I_{\mathrm{S}}(T)$ dependence of the V type is also characterized by the existence of a compensation temperature T_{comp} and does not differ principally from the N type dependence.

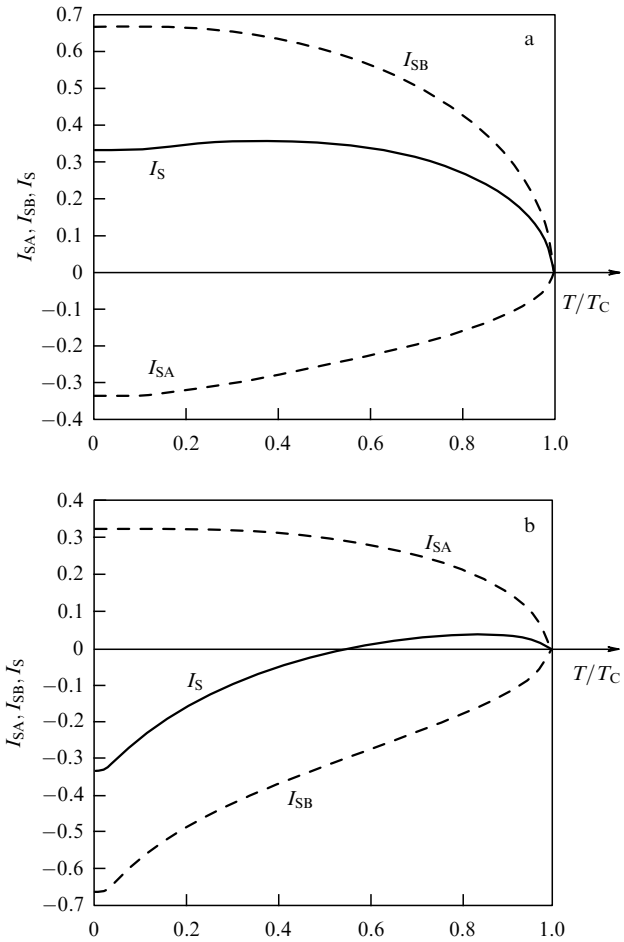

Figure 5. (a) Temperature dependences of spontaneous magnetizations I_{SA} and I_{SB} of the ferrimagnetic A and B sublattices and of the total spontaneous magnetization I_{S} of a ferrimagnet of the Néel P type ($\alpha = 1$, $\beta = -0.6$, $\lambda = 1/3$, and $\mu = 2/3$). (b) Temperature dependences of the spontaneous magnetizations I_{SA} and I_{SB} of the ferrimagnetic A and B sublattices and of the total spontaneous magnetization I_{S} of a ferrimagnet of the Néel N type ($\alpha = 1.26$, $\beta = -0.39$, $\lambda = 1/3$, and $\mu = 2/3$).

The change of sign in I_{S} and, correspondingly, in the TRM and CRM, as was mentioned above, can occur in relatively weak magnetic fields as a result of both different temperature dependences of the spontaneous magnetizations of individual sublattices in the ferrimagnet and diffusion of magnetic ions or point defects between the sublattices that occurs as time goes on. In both the cases (N, V), there is considered self-reversal related to the only mineral phase and, therefore, we speak of a single-phase physical mechanism which we will further call the Néel N type mechanism or simply N type mechanism.

There also exist other concepts concerning the mechanisms of self-reversal of magnetization in ferrimagnetic minerals of rocks [4, 11]. It was shown as long ago as in 1954 that the self-reversal of magnetization can occur due to magnetostatic interactions between two contacting samples of two minerals [26]. It is assumed that the magnetostatic interaction is also possible in natural fine ferrimagnetic grains possessing a two-phase state [11]. The possibility of self-reversal of two-phase minerals due to exchange interaction between different phases was also considered in Ref. [11].

As appears from the above, all physical mechanisms of self-reversal of magnetization in ferrimagnets can, in principle, be classified into single-phase and multiphase (as a rule, two-phase). In the case of two-phase mechanisms, the self-reversal is caused by the interaction of mineral phases between each other.

There are only scarce theoretical works concerning self-reversal of magnetization, and as basic works we can only consider the above-cited works by Néel [20–22]. In the work published in 1951 [20], Néel applied the theory of collinear ferrimagnetism to rocks and suggested all four above-mentioned mechanisms of self-reversal of magnetization. It should be noted that in two-phase mineralogical systems, according to Néel, upon the self-reversal of magnetization of rocks resulting from the interaction of two phases, the magnetostatic interaction takes place in the entire volume of the interpenetrating phases, whereas the exchange interaction shows up only at the interphase boundary.

The single-phase mechanisms of self-reversal have already been discussed; now, we consider in more detail two-phase mechanisms [20, 27]. Let the phases A and B be characterized by different Curie temperatures T_{CA} and T_{CB}, and let us assume that $T_{CA} > T_{CB}$. Then, the magnetization of the two-phase system can be written down as

$$I(T) = C_A I_A(T) + C_B I_B(T)\,, \tag{3.6}$$

where C_A and C_B are the volume concentrations of the phases A and B, respectively.

When the total TRM is developed in a weak field H for $T_{CA} > T_{bA} > T_{CB}$, the magnetization induced by the field can be represented as

$$I(T) = C_A I_A(T) = C_A \lambda_A(T_{bA})\, I_{SA}(T)\, H\,, \tag{3.7}$$

where $\lambda_A(T)$ is the coefficient of proportionality, and T_{bA} is the temperature of blocking the phase A. If at a temperature less than or equal to T_{bA}, the $I_A(T)$ magnetization is completely blocked, the interaction between the A and B phases in the above temperature range will exert influence only on $I_B(T)$.

With allowance for the interaction between the phases, the magnetization for $T < T_{bA}$ can be written as

$$\begin{aligned} I(T) = {} & C_A \lambda_A(T_{bA})\, I_{SA}(T)\, H \\ & + C_B \lambda_B(T_{bB})\, I_{SB}(T)\, \big[H - N I_A(T)\big]\,, \end{aligned} \tag{3.8}$$

where $[H - NI_A(T)]$ and T_{bB} are the effective field and the blocking temperature, respectively, for the B phase. The demagnetizing factor N depends on the geometry of the phases and on the nature of their interaction. As $N > 0$, the interaction is negative and the condition for reversal of $I_B(T)$ has the form

$$\frac{H}{I_A(T)} = \frac{1}{C_A \lambda_A(T_{bA})\, I_{SA}(T)} < N\,, \tag{3.9}$$

while the condition for the reversal of magnetizations of the entire system (3.6) is expressed as

$$\frac{1}{C_A \lambda_A(T_{bA})\, I_{SA}(T)} + \frac{1}{C_B \lambda_B(T_{bB})\, I_{SB}(T)} < N\,. \tag{3.10}$$

Notice that the magnetostatic interaction in the two-component model is by no means always negative.

When the self-reversal of magnetization is caused by the Weiss–Heisenberg exchange interaction at the phase boundary, a consistency is required between the crystal lattices of both components; the magnitude of the exchange field inducing self-reversal can be around that of the molecular field (of the single-component Néel model).

3.2 Ferrimagnetic mineralogy

Before we begin our review of the literature concerning the problem of self-reversal of magnetization in natural and synthesized ferrimagnets that is observed in the laboratory conditions in rock samples, it is important to pay attention to their chemical composition. Therefore, we dwell in more detail on the magnetic mineralogy of rocks [10, 27].

The minerals that are constituents of rocks can be dia-, para-, ferro-, ferri-, or antiferromagnetic. Quartz and feldspar are diamagnetic minerals; olivines, pyroxenes, and biotites are paramagnetic. The ferrimagnetism of rocks is related to various minerals of the oxide group present in them, which contain Fe^{2+} or Fe^{3+} as the main components (e.g., magnetite, hematite, ilmenite, etc.) and possess ferrimagnetic or antiferromagnetic ordering; their ferromagnetism is due to native iron.

Some oxides form continuous solid solutions. The most common in rocks are solid-solution series of the magnetite–ulvospinel (titanomagnetites) and hematite–ilmenite (hemoilmenites) systems, as well as iron hydroxides. All the other magnetically ordered minerals occur on the whole much less frequently than the ferrospinels, sulfides, native iron and some others. Correspondingly, their contribution to the magnetic characteristics of rocks is very small. Now, we briefly consider the main characteristics of the carriers of magnetization in rocks.

3.2.1 Ferrospinels and titanomagnetites. A large group of minerals of the cubic system have a structure of the mineral called spinel ($MgOAl_2O_3$). The unit cell of the ferrospinel is a cube consisting of 32 oxygen anions between which there are located 64 tetrahedral (A) and 32 octahedral (B) interstices being partially filled with cations such as Fe^{2+}, Mg^{2+}, Mn^{2+}, Zn^{2+}, Ni^{2+}, Co^{2+}, Fe^{3+}, Al^{3+}, Cr^{3+}, Mn^{3+}, Ti^{4+}, or Mn^{4+}. The general formula of the spinel is written down as $M_x^{2+}\,Fe_{1-x}^{3+}\big[M_{1-x}^{2+}Fe_x^{3+}\big]\,O_4$ (ions occupying octahedral positions are indicated in brackets; M stands for a metal atom). In titanomagnetites, $2Fe^{2+}$ cations are replaced by $Fe^{2+} + Ti^{4+}$.

Titanomagnetite constitutes a solid solution of magnetite (Fe_3O_4) and ulvospinel (Fe_2TiO_4). The ferrospinels frequently occur in magmatic and metamorphic rocks. Titanomagnetite is encountered in the overwhelming majority of continental and ocean magmatic rocks which sometimes form large deposits. These rocks are of large practical importance. The chemical formula of titanomagnetite as a solid solution of magnetite and ulvospinel is written down as $xFe_3O_4(1-x)\,Fe_2TiO_4$.

Synthesized titanomagnetites (from magnetite to ulvospinel) were for the first time prepared by Japanese researchers

Akimoto, Katsura, and Yosida [28]. As high-temperature formations, the titanomagnetites are most frequently encountered in magmatic rocks and their composition in such rocks usually varies from $Fe_{2.2}Ti_{0.8}O_4$ to $Fe_{2.3}Ti_{0.2}O_4$, neglecting impurities (see Buddington, Lindsley [29]).

Apart from titanium, the titanomagnetites frequently contain magnesium, manganese, calcium, aluminum, chromium, vanadium, etc., whose total content in effusive rocks does not exceed usually 10%.

Titanomagnetite is metastable and it is retained in rocks only upon rapid cooling. Beginning from approximately 300 – 400 °C, titanomagnetite is oxidized in air and is decomposed with the formation of a magnetite aggregate or a titanomagnetite of an approximate composition and intergrowths [usually along (111) planes] of hemoilmenite, which at a later time is oxidized to minerals of the pseudobrookite series ($Fe_2TiO_5 - FeTi_2O_5$) and to rutile (TiO_2); at temperatures below 300 – 400 °C, titanomagnetite can oxidize (the oxidation usually proceeds in the presence of water), retaining its single-phase state and forming defect metastable titanomagnetites which are called titanomaghemites (the defect structure is due to vacancies that arise in the oxidation process $Fe^{2+} \to Fe^{3+}$; see, e.g., Ref. [30]). The process of single-phase oxidation of titanomagnetite is observed in virtually all ocean basalts. In the course of low-temperature oxidation with the formation of titanomaghemites, a constant ratio $Fe/Ti \geqslant 2$ of the iron content to titanium content is retained, but the Fe^{3+}/Fe^{2+} ratio varies. The defect (vacancy-containing) structure of titanomaghemites is unstable; upon heating, the titanomaghemites are easily decomposed with the formation of a magnetite aggregate or a titanomagnetite of an approximate composition and intergrowths of hemoilmenite, which is frequently close in composition to ilmenite.

The magnitude of the magnetic moment of titanomagnetite depends on the ratio between Fe^{2+} and Fe^{3+}, i.e., on the degree of oxidation. During oxidation, the Fe^{2+} ions occupy octahedral positions (prior to oxidation, they were located in both octahedral and tetrahedral positions); as a result, an increase in the uncompensated part of the magnetic moment of the B sublattice is observed. This fact, as well as an increase in the degree of ordering of the structure and a decrease in the dimensions of the unit cell of titanomagnetite, leads to an increase in both I_S and T_C. The appearance of vacancies upon the formation of titanomagnetite, on the contrary, leads to a decrease in I_S, The coercive force upon titanomagnetite oxidation has a tendency to increase because of the destruction of the homogeneous grain and appearance of a macrostructure, internal stresses, and dislocations.

Judging from the results of heat treatments, only a single primary phase (predominantly, titanomagnetite) is contained in all igneous rocks. Multiphase variants are mainly related to oxidation, decomposition, and, correspondingly, incomplete homogenization upon heat treatment rather than to differences in the processes of generation of magnetic minerals.

3.2.2. Solid solutions of hemoilmenites. Hematite α-Fe_2O_3, being one of the terminal components of the $xFeTiO_3\,(1-x)\,Fe_2O_3$ series of solid solutions [31], is widespread in nature and is a canted antiferromagnet with a very small I_S. The Curie temperature of hematite is $T_C \approx 700$ °C. The Néel temperature of pure ilmenite ($FeTiO_3$), the other terminal component of these solid solutions, is equal to approximately 68 K, while that of hematite is 948 K. The melting point of hematite lies in the range of 1700–1800 °C, and that of ilmenite is 1470 °C.

The minerals of the hemoilmenite series, which represent solid solutions of hematite and ilmenite, have a rhombohedral structure with lattice parameters corresponding to a chemical composition $xFeTiO_3(1-x)Fe_2O_3$ and varying between a value of $a = 5.42$ Å characteristic of hematite and $a = 5.54$ Å corresponding to ilmenite [32]; their Curie temperatures also lie, depending on the composition, between the Curie temperatures of hematite and ilmenite. The minerals of the ilmenite – hematite series for $1 > x \geqslant 0.45$ are ferrimagnetic, while those with $0.5 > x \geqslant 0$ are antiferromagnets with a weak superimposed ferromagnetism. The samples of solid solutions corresponding to the area of weak ferromagnetism possess, as compared to ferrimagnetic differences, a higher coercive force and small remanent magnetization.

The solid solutions revealing ferrimagnetism at room temperature and, therefore, determining the magnetic properties of rocks are restricted to a narrow composition range of $0.45 \leqslant x < 0.8$. Some solid solutions in this range possess the ability of acquiring a reversed (self-reversed) thermoremanent magnetization. Uyeda [31] specified this composition range as $0.45 < x < 0.6$. Samples with a composition of $0.45 \leqslant x \leqslant 0.5$ immediately after synthesis at $T = 1200$ °C and subsequent cooling exhibited only direct thermoremanent magnetization, and only after a repeated heat treatment (with a slow cooling from $T = 1000$ °C) did they become magnetized in the opposite direction.

The ability of hemoilmenites to acquire reverse magnetization, when their composition approaches the boundary between the ferrimagnetic and antiferromagnetic compositions, is a distinguishing feature of hemoilmenites.

3.3 Self-reversal of the magnetization of hemoilmenites

In 1951, after the publication of Néel's work [20], Nagata et al. [33, 34] revealed self-reversal of the thermoremanent magnetization of igneous rocks (dacites) from Haruna Mountain, which contained hemoilmenites. It is traditionally assumed that this is the first well-documented evidence of self-reversal of magnetization of rocks. Therefore, the works of Nagata and colleagues of 1951 [33, 34], along with Néel's work of the same year [20], are also key publications for researchers of self-reversal. Later on, even a special term was introduced, namely, a 'Haruna type' self-reversal of magnetization. However, Kneller indicates in his monograph *Ferromagnetismus* (*Ferromagnetism*) [35] that the self-reversal of remanent magnetization of a sample of carbonyl iron had already been observed in 1922. In subsequent years, the self-reversal of magnetization of samples from Haruna Mountain was repeatedly reproduced by various researchers [36 – 38].

Since the revelation (1951) of the existence of the physical mechanisms of the formation of a reverse natural remanent magnetization, i.e., of mechanisms of self-reversal of magnetization of rocks, and since the self-reversal of magnetization was experimentally examined in the laboratory, there arose a question of the physical mechanism of formation of the reverse NRM in each concrete case where it was revealed in nature: whether this is a result of an opposite direction of the GMF or the effect of one of the mechanisms of self-reversal. Therefore, in each case where the reverse NRM was subsequently revealed, attempts were made to clarify the concrete cause of its formation.

In 1951, after the publication of the works by Néel [20] and Nagata et al. [33, 34], Roche [39] studied 35 lavas from

the mountains of the Auvergne province of France and revealed that some of the samples possessed a reverse NRM. Initially, Roche assumed that the reverse NRM could be formed as a result of both GMF reversals and self-reversal of magnetization. In 1953, he performed a number of additional experiments on samples with a reverse NRM in order to clarify the source of its formation [40]. The experiments on thermodemagnetization, demagnetization in an alternating magnetic field h (50 Hz), and thermomagnetization in the GMF did not disclose properties of self-reversal in the samples studied; therefore, Roche concluded that the formation of reverse magnetization as a result of self-reversal was unlikely. Notice that the above experiments are by no means always sufficient to expose the properties of self-reversal; frequently, more detailed experimental investigations are required (see Section 4.2).

At the same time, Nagata was faced with the question of clarifying the physics of the self-reversal phenomenon, i.e., of the determination of a plausible physical mechanism responsible for the self-reversal of magnetization in the samples of dacitic pumices from Haruna Mountain. In succeeding years, Nagata continued studying self-reversal in samples from the mountain [27, 41–45]. He also proposed a hypothesis for a physical mechanism of self-reversal of magnetization in hemoilmenites of dacitic pumices, which was subsequently refined and detailed by other researchers (see Section 4). However, as was mentioned in Section 3.2, ferrimagnetic minerals of rocks have a very complex composition and, as a rule, contain several minerals or mineral phases, so that no common opinion about the mechanism of self-reversal of historical lavas from Haruna Mountain have been accepted to date.

The conclusion made by Uyeda [31] to the effect that it is only hemoilmenites $x\mathrm{FeTiO_3}(1-x)\,\mathrm{Fe_2O_3}$ with an ilmenite molecular fraction x in the range of $0.45 \leqslant x \leqslant 0.6$ that can be the carriers of the reverse thermoremanent magnetization (TRM), i.e., can possess properties of self-reversal, was confirmed by numerous experiments by various researchers [31, 38]. In 1958, when studying samples of dacitic pumices from Haruna Mountain containing hemoilmenites with $x \approx 0.5$, Uyeda again examined self-reversal. Later on, in 1962–1963, Ishikawa and Syono [46, 47] investigated the self-reversal effect using synthesized hemoilmenites (also with a composition of $x \approx 0.5$). They also constructed a first phenomenological model of self-reversal of magnetization (see Section 4). In 1965, Petrova et al. [48] observed self-reversal in hemoilmenites from the Kamchatka peninsula, Russia. As is seen from the above facts and evidenced by the ensuing (see Sections 3.4 and 3.5), the self-reversal of magnetization of natural and synthesized hemoilmenites of a certain composition has been obtained repeatedly at various points on the Earth, for example, in synthesized ilmenite–hematite oxides [49], synthesized hemoilmenites [50], and so forth.

In 1984–1985, Trukhin et al. [51] for the first time found self-reversal in samples from kimberlite diamond pipes and in samples of traps from Yakutia, Russia. In kimberlites, the property of self-reversal was exhibited by picroilmenites being the analogs of hemoilmenites. In 1997, Trukhin et al. [1] observed self-reversal in synthesized hemoilmenites of various compositions (see Fig. 6). As can be seen from Fig. 6, the compensation points were located in a temperature range of 80–200 °C, and the concentration of ilmenite x varied from 0.55 to 0.62.

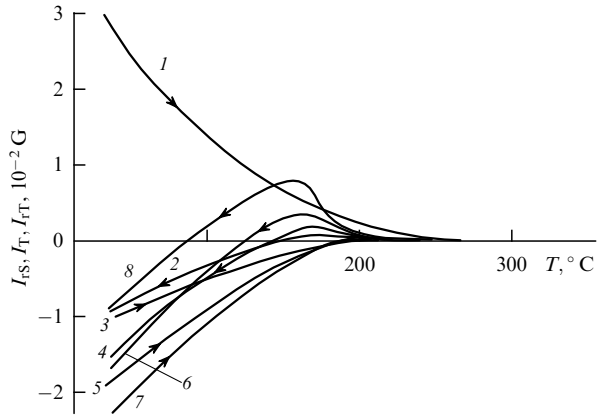

Figure 6. Curves of the temperature dependence of the remanent saturation magnetization $I_{rS}(T)$, thermomagnetization $I_T(T)$, and thermoremanent magnetization $I_{rT}(T)$ of a sample of the synthesized hemoilmenite $x\mathrm{FeTiO_3}(1-x)\mathrm{Fe_2O_3}$ with a molecular fraction of ilmenite $x = 0.55$: (*1*) $I_{rS}(T)$ curve (heating in a field of $H = 0$ to $T = 425\,°\mathrm{C}$); (*2*) $I_T(T)$ curve (cooling in a field of $H = 2.5$ G); (*3*) $I_{rT}(T)$ curve (heating in a field of $H = 0$ to $T = 425\,°\mathrm{C}$; (*4*) $I_T(T)$ curve (cooling in a field of $H = 10$ G); (*5*) $I_{rT}(T)$ curve (heating in a field of $H = 0$ to $T = 380\,°\mathrm{C}$; (*6*) $I_T(T)$ curve (cooling in a field of $H = 25$ G); (*7*) $I_{rT}(T)$ curve (heating in a field of $H = 0$ to $T = 390\,°\mathrm{C}$), and (*8*) $I_T(T)$ curve (cooling in a field of $H = 50$ G) [1].

Notice separately two more cases of self-reversal in hemoilmenites: Haag et al. [52] in 1990 obtained self-reversal in andesitic pumice from the eruption of a volcano of the Nervado del Ruiz (Colombia, 1985), and Ozima et al. [53] in dacite pumice from the eruption of the Pinatubo volcano (Philippines, 1991). Samples of the same eruption of the Nervado del Ruiz volcano were later studied by other researchers (see, e.g., Refs [54, 55]). Samples from the eruption (1991) of the Pinatubo volcano have also been studied repeatedly [56–58].

3.4 Self-reversal of the magnetization of titanomagnetites

Notice that self-reversal of magnetization which, according to some authors [31, 38], is an inherent property of hemoilmenites has been observed not only in hemoilmenites. In 1965, Havard and Lewis [59] detected self-reversal of TRM in the samples of titanomagnetite-containing basalts from India. In 1966, O'Reilly and Banerjee [60] exposed this effect in samples of synthesized titanomagnetites. Apart from continental basalts, self-reversal properties can also be characteristic of submarine basalts. For example, in 1968, Ozima and Larson [61] revealed self-reversal of TRM in samples of oceanic basalts from various parts of the Pacific Ocean. The samples contained titanomagnetites, as in the case of continental basalts.

Later on, self-reversal of magnetization was repeatedly observed in samples of continental and oceanic (submarine) basalts containing titanomagnetites and/or titanomaghemites [2, 62–65], as well as on synthesized titanomagnetites of composition $\mathrm{Fe_{2.4}Ti_{0.6}O_4}$ [66]. Self-reversal was also registered in synthesized magnesian titanomagnetites $\mathrm{Fe}^{3+}_{2-2t}\mathrm{Fe}^{2+}_{1+t-m}\mathrm{Mg}_m\mathrm{Ti}_t\mathrm{O_4}$ for $0.25 \leqslant m \leqslant 0.5$, and $0.5 \leqslant t \leqslant 1$ [67], where m is the content of magnesium, and t is the content of titanium. The compensation points T_{comp} of magnesian titanomagnetites lie in the range from -50 to $+200\,°\mathrm{C}$.

In 1980, Heller [68] for the first time obtained self-reversal in titanomagnetite samples from Laschamp and Olby

(France). Previously, it was assumed that the reverse NRM of these samples is a consequence of magnetization of the rocks in the GMF of the opposite sign, which existed at the time of the GMF excursion. Later, samples of this deposit were studied repeatedly (for details, see, e.g., Ref. [69]).

The self-reversal of magnetization that was observed in Refs [51, 70] on samples of traps from Yakutia was caused by self-reversal of magnetization of titanomagnetites contained in these traps.

Recently, a number of works on the self-reversal of magnetization of titanomagnetites and titanomaghemites from submarine oceanic basalts have appeared [2, 71, 72]

3.5 Self-reversal of magnetization of other minerals

Self-reversal of magnetization has been obtained not only in titanomagnetites and hemoilmenites. In 1954, when studying some ferrites with the purpose of confirming Néel's theory [22], Gorter [73] revealed self-reversal of magnetization in chromium – lithium ferrite; it turned out that only ferrite of a certain composition exhibited properties of self-reversal. This case of self-reversal is historically considered one of the first to be revealed, but it fundamentally differs from the case of self-reversal found in the dacites of Haruna Mountain, since it was observed in a sample which consisted completely of a pure synthesized ferrimagnet rather than in a sample of a rock, so that the magnetic signal from such a sample was much stronger than that of a rock sample.

In 1954, Grabovskii and Pushkov [26] observed self-reversal in samples consisting of tightly joined magnetite and pyrrhotite. In 1968, Kropacek [74] obtained self-reversal in natural cassiterite. In 1973, Bol'shakov et al. [75] revealed self-reversal of magnetization in samples of magnetite from the Kovdor magnetite deposit (Kola peninsula, Russia).

Self-reversal was also exposed in crystals of pyrrhotite [76, 77] and, in 1993, in synthetic lepidocrocites containing maghemite [78].

Later on, andesitic pumices from Sakura-jima Mountain (Japan), which possess the property of self-reversal, were studied [36, 79]. An analysis of the composition of these samples showed the presence of titanomagnetite and chromite, the latter in some cases being oxidized to magnetite.

Mention should also be made of self-reversal of magnetization that was obtained in samples of Berlin blue $(Ni_{0.22}Mn_{0.60}Fe_{0.18})_{1.5}[Cr(CN)_6]\times 7.6H_2O$ and in nickel salt of formic acid, $Ni(HCOO)_2 \times 2H_2O$, in 2004 [80]. Historically, self-reversal was for the first time obtained in nickel as long ago as in 1888 (see, e.g., Ref. [61]). However, since nickel is not a ferrimagnet, this work, just as that by Gorter [73], has been ignored, as a rule, in the literature on rock magnetism. Nevertheless, the clarification of the mechanism of self-reversal of nickel is also of great scientific interest.

As is seen even from this brief review, the self-reversal of magnetization is a rather widespread phenomenon in nature and frequently encountered in synthesized ferrimagnets.

3.6 Peculiarities of the self-reversal effect

The experiments carried out by the majority of authors mentioned in Sections 3.1 – 3.5 show that it is precisely samples with a reverse NRM that possess the property of self-reversal of NRM or TRM [in some works, self-reversal of IRM (see, e.g., Ref. [76]) and CRM (see, e.g., Ref. [78]) is also considered]. The authors of Refs [52, 81] state with confidence that the reverse NRM of their samples was formed in nature as a result of the process of self-reversal of magnetization rather than of GMF reversals in past geological epochs. They arrived at such a conclusion based on the experimental examination of a virtually complete coincidence of the thermodemagnetization curves for NRM and TRM, in which the magnetization changes its sign upon passage through the compensation temperature.

It should be emphasized that the self-reversal effect can be observed in the curves of both NRM thermodemagnetization and thermomagnetization in a field of $H > 0$ (see Fig. 6) or TRM thermodemagnetization at $H = 0$ (see Fig. 6). As an illustration of the self-reversal effect, different curves of the temperature-dependent magnetization (TDM) are usually given in various works; however, most frequently the change in the sign of magnetization of a sample is observed in the curves of TRM thermodemagnetization.

The self-reversal of magnetization can be 'complete' or 'partial'. Complete self-reversal is the effect in which the magnetization of a rock sample changes its sign when the temperature, while changing, passes through the compensation point (see Fig. 6). By partial self-reversal is meant an anomalous decrease in the curve of laboratory thermomagnetization or, on the contrary, an increase in the magnetization upon laboratory thermodemagnetization of the TRM or NRM of the sample, which are not accompanied by a further change in the magnetization sign (Fig. 7, curves *1*, *2*).

Apart from the complete thermoremanent magnetization, there also exists partial thermoremanent magnetization (PTRM) I_{rpT}, which can also offer self-reversing properties (Fig. 8) [2]. Partial thermoremanent magnetization, like TRM, arises during the cooling of a sample in a magnetic field with the only difference that the field is switched on in a certain temperature range (from a few to several hundred kelvins) rather than in the entire temperature range $[T_C, T_0]$, where T_C is the Curie point, and T_0 is room temperature. After the sample acquires a PRTM, it can exhibit properties of self-reversal during further cooling in the absence of a field; it is this phenomenon that is called self-reversal of the PRTM.

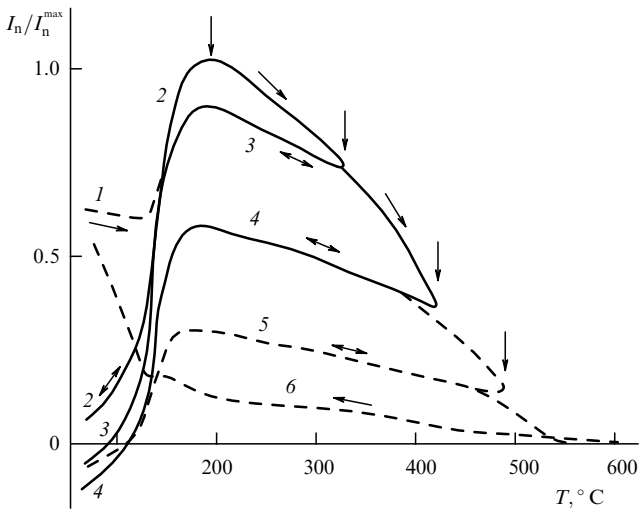

Figure 7. Cyclic thermodemagnetization ($H = 0$) of the natural remanent magnetization (NRM) I_n of a sample of kimberlite (Mir diamond pipe, Yakutia) containing ferrimagnetic picroilmenites (natural analogs of hemoilmenites): (*1*) heating to $T = 100\,°C$; (*2*) cooling to $T_0 \approx 20\,°C$ and heating to $T = 320\,°C$; (*3*) cooling to T_0 and heating to $T = 420\,°C$; (*4*) cooling to T_0 and heating to 490 °C; (*5*) cooling to T_0 and heating to 600 °C, and (*6*) formation of a thermoremanent magnetization I_{rT} on cooling from $T = 600\,°C$ to T_0.

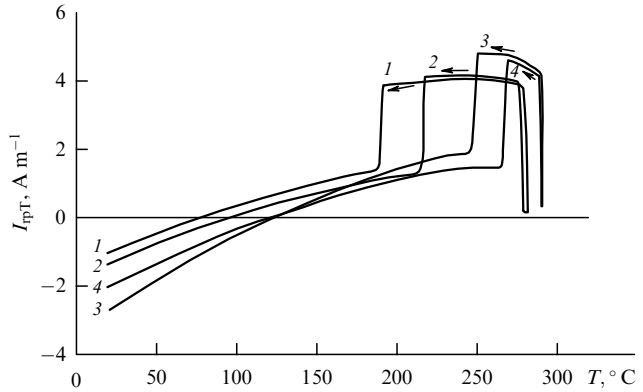

Figure 8. Curves of the formation of partial thermoremanent magnetization (PTRM) I_{rpT} of a sample of basalt (taken from the bottom of the Romanche transform fault, Central Atlantic) containing a titanomagnetite $x\mathrm{Fe_3O_4}(1-x)\mathrm{Fe_2TiO_4}$: (*1*) cooling in a field of $H = 0.5$ mT from $T = 280$ to 200 °C, then cooling in a zero field; (*2*) cooling in a field of 0.5 mT from $T = 280$ to 235 °C, then cooling in a zero field; (*3*) cooling in a field of 0.5 mT from $T = 290$ to 250 °C, then cooling in a zero field, and (*4*) cooling in a field of 0.5 mT from $T = 290$ to 270 °C, then cooling in a zero field.

Thus, the class of minerals that possess the property of self-reversal of magnetization is quite narrow: these are mainly hemoilmenites, titanomagnetites, and their analogs. Notice that in magma which cools after eruption onto the Earth's surface the primary crystallizing minerals are titanomagnetites. Other ferrimagnetic minerals, in particular, hemoilmenites, appear as a rule as a result of subsequent chemical transformations. It is pertinent also to note that self-reversal has mainly been observed in igneous rocks. Sedimentary rocks, which are formed as a result of transformations and re-deposition of igneous rocks, have not been studied so widely as igneous ones. Experimentation using sedimentary rocks is fraught with significant difficulties, since they are subject to chemical transformations upon heating to high temperatures and subsequent cooling in a magnetic field. Nevertheless, self-reversal of magnetization in sediments was obtained in Ref. [48]; therefore, sediments can also possess properties of self-reversal and the problem of the self-reversal of magnetization of sedimentary rocks is topical now. We emphasize once again that sedimentary rocks are also carriers of the reverse NRM in nature. Sedimentary rocks have more widely been used in paleomagnetic investigations than have igneous ones [4].

As is seen from the above historical review, the samples of rocks that offer properties of self-reversal of magnetization were selected from a very limited number of places (Fig. 9). As will be shown in Section 4, no common viewpoint concerning the self-reversal mechanism has been accepted to date. However, the two-phase mechanisms have a greater number of advocates. This, apparently, can also be explained by the 'narrowness of the geography': the most frequently studied samples were taken from the igneous rocks of Haruna Mountain and other volcanoes of Japan, as well as from lavas of Pinatubo Mountain (Philippines) and Nervado del Ruiz (Colombia). It is worth noting that by no means do all of the works on self-reversal claim to have determined the concrete physical mechanism responsible for this phenomenon. The commonly accepted viewpoint on the self-reversal mechanism would possibly change after studying self-reversing samples from other geographic points on the Earth. This has already been done partly (see Fig. 9), but the main discussion on the self-reversal mechanisms has so far persisted around the above-mentioned geographic places. Hence the 'skewness' toward two-phase mechanisms of self-reversal.

Several research teams around the world are now engaged in studying the effect of self-reversal of magnetization of rocks, for example, those of Trukhin (Russia) and Ozima (Japan), along with Dunlop (Canada), Heller (Germany), Krása (Germany), Petersen (Germany) and some others.

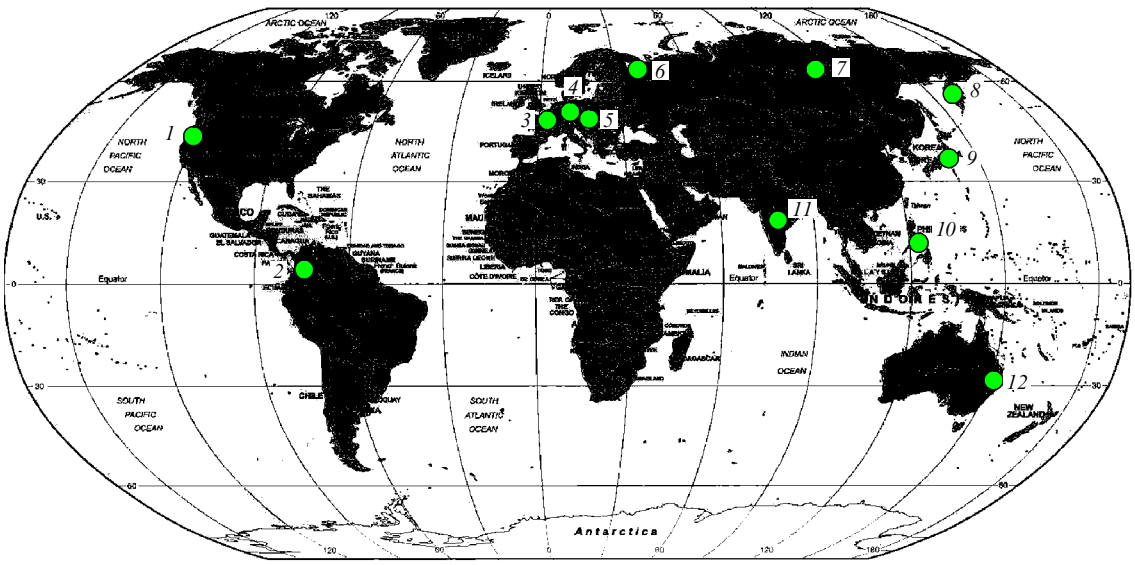

Figure 9. Map of the world. Circles indicate places where samples that exhibit properties of self-reversal in laboratory studies were taken: (*1*) United States (northern California, Mount Shasta); (*2*) Colombia (Nervado del Ruiz volcano); (*3*) France (Laschamp and Olby, province Auvergne); (*4*) Germany (Fogelsberg); (*5*) Central Slovakia (Stiavnica, Javori, Pol'ana mountains [82]; (*6*) Russia, Kola peninsula (Kovdor magnetite deposit); (*7*) Russia, Yakutia; (*8*) Russia, Kamchatka peninsula; (*9*) Japan (Haruna, Akagi, Sambre, and other volcanoes); (*10*) Philippines (Pinatubo, Natib Mountains); (*11*) India, and (*12*) Australia (New South Wales Mountain) [83].

Each research group and each individual researcher have, as a rule, their own viewpoint on the mechanism of self-reversal of magnetization and hold to their views when investigating various samples of rocks.

4. Possible physical mechanisms of experimentally established cases of self-reversal and a model of the phenomenon

As was already mentioned above, rocks have a very complex composition. A rock usually represents a matrix consisting of dia- and paramagnetic minerals with imbedded ferrimagnetic minerals in the form, as a rule, of fine grains. The grains can be single-domain, pseudo-single-domain, or multidomain, and can exhibit uniaxial or triaxial magnetic anisotropy. Ferrimagnetic minerals in the form of uniaxial (shape anisotropy) single-domain ferrimagnetic grains are frequently encountered in rocks. The grains can also be single-phase or multiphase, i.e., consist of one or several mineral and/or magnetic phases. The Curie points of various phases are, as a rule, different.

The ferrimagnetic grains that enter into a rock can in turn have a complex structure; in addition, a rock can contain ferrimagnetic inclusions of various chemical compositions. Thus, a rock represents a conglomerate of compositionally nonuniform minerals, and the exact chemical composition of a concrete sample of a rock only rarely is known exactly. It is mainly for this reason that a common viewpoint on the self-reversal mechanism is absent, and the revealing of a mineral (minerals) responsible for the self-reversal in each concrete case is in itself a very complex problem.

Reviews on the problem of self-reversal of magnetization can be found in the monographs [3, 4, 11, 84], of which the most detailed is Ref. [4]. In our review, we consider the most interesting, in our opinion, works on self-reversal of those that have been published.

4.1 Mechanisms of self-reversal of magnetization of natural and synthesized ferrimagnets

We first consider in a chronological order models of self-reversal of magnetization of natural ferrimagnets. Note here that all the models to be discussed are purely empirical and phenomenological, based on the two-phase mechanisms of self-reversal suggested by Néel in 1951.

In 1954, Grabovskii and Pushkov [26] for the first time showed experimentally that a magnetostatic interaction of two phases can indeed lead to a self-reversal of magnetization that can be observed in the laboratory. They tightly joined two ferrimagnets, namely, magnetite and pyrrhotite. The Curie point of magnetite is 585 °C, and that of pyrrhotite is 350 °C. Upon cooling down from 585 °C to 350 °C in a magnetic field of 1 G, there occurred first a magnetization of the magnetite; then, in the demagnetizing field of the magnetite, which was antiparallel to the applied magnetic field and exceeded it in magnitude, the pyrrhotite became magnetized. As a result, the total magnetization of the two samples acquired a direction antiparallel to the magnetizing field. In this experiment, it was reliably established that self-reversal of magnetization occurred as a result of the action of a two-phase mechanism.

The first case of self-reversal of thermoremanent magnetization of a rock sample, which was observed by Nagata in the laboratory in hemoilmenites from dacite pumices taken from Haruna Mountain, the researcher ascribed to both magnetostatic [27] and exchange [45] interactions at the boundary between two hemoilmenite phases.

Uyeda [31, 85], who investigated hemoilmenite-containing samples of igneous rocks from Haruna Mountain and observed their self-reversal, also explained the self-reversal of magnetization of his samples by an exchange interaction at the boundary of two hemoilmenite phases with the same chemical composition but with different Curie points. The first phase is, according to Uyeda [31, 85], an ordered ferrimagnetic phase (FiM) with a lower T_C. The second phase is a disordered antiferromagnetic phase with a weak ferromagnetism (WF); it possesses a higher T_C. Upon cooling, the WF phase, whose magnetization occurs first because of its higher Curie temperature, becomes magnetized in the direction of the applied magnetic field. Upon further cooling, the FiM phase becomes magnetized antiparallel to the external magnetic field as a result of negative exchange interaction with the WF phase (the energy of this interaction is greater by several orders of magnitude than that of the applied magnetic field). The carrier of the reverse magnetization is the ordered ferrimagnetic hemoilmenite phase with a molecular fraction of ilmenite $x \approx 0.5$; if, as a result of cooling to room temperature T_0, the direct component of the magnetization becomes less than the reverse component, the sample turns out to have a reverse magnetization. Uyeda does not state that this mechanism of self-reversal of magnetization is universal and applies it only to explain self-reversal in hemoilmenite-containing dacites of Haruna Mountain. Haag et al. [52, 55, 86] also employ this model to explain self-reversal of magnetization of andesitic pumices from the eruption of the Nervado del Ruiz volcano (Colombia, 1985), but they state that the two interpenetrating hemoilmenite phases (FiM and WF), whose exchange interaction is responsible for the self-reversal, have slightly different chemical compositions. The self-reversal effect can be suppressed because of the process of the formation of a domain wall inside the reversely magnetized phase. With increasing field, the wall moves toward the phase boundary and at a high field strength the directly magnetized domain will suppress the reversely magnetized domain.

Ishikawa [87] noted that Uyeda's conclusion [31, 85] on the interaction mechanism of two hemoilmenite phases, which causes the formation of a reverse TRM, is not true since, as follows from experimental data, if both phases have the same composition, the Curie point of the ordered phase should be higher than that of the disordered phase. On this basis, Ishikawa and Syono [46, 47] introduced the so-called x phase and suggested a three-phase model of self-reversal of magnetization.

Let us consider this three-phase model of self-reversal [46, 47] in some more detail. As will be shown below, this model is widely used for the explanation of self-reversal of magnetization in various igneous rocks. The authors of Refs [46, 47] have studied in detail the order – disorder transition and the reverse TRM of synthesized solid solutions of hemoilmenites $x\mathrm{FeTiO_3}(1-x)\mathrm{Fe_2O_3}$ with $x \approx 0.5$ and concluded that the reverse TRM is tightly connected with the order – disorder transition in hemoilmenites. In this case, the reverse TRM is only observed in samples that are in a state of metastable equilibrium rather than in the completely ordered or completely disordered samples. Apart from two hemoilmenite phases, FiM and WF, the authors of Refs [46, 47] introduce a third metastable phase, the so-called x phase, which is formed around the ordered phase in the course of ordering and then

disappears. According to their model, the Curie temperature T_C of the metastable phase is higher than that of the FiM phase; therefore, the metastable phase becomes magnetized first, exhibiting magnetization in the direction of the magnetizing field. The reverse TRM is formed as a result of a negative exchange interaction between the magnetic moments of the ordered phase and moments of the iron-enriched metastable phase, or x phase, which is formed around the ordered structure in the course of ordering. Ishikawa and Syono emphasize that the properties of the reverse TRM, such as, for example, its intensity, are closely connected with the conditions of cooling, e.g., cooling duration and annealing temperature.

The model of self-reversal of magnetization suggested by Ishikawa and Syono in 1963 has long had no analogs; it was used and is used by many researchers for the explanation of self-reversal of magnetization in samples of igneous rocks containing hemoilmenites or titanohematites [38, 54, 86].

The above model has one weak point — its authors do not indicate the exact composition of the weakly magnetic x phase, and nothing is said of its nature, geometry, or how this phase is formed in the hemoilmenite crystal. Therefore, later attempts to refine this model and to explain what the x phase is in reality have been undertaken in many works. However, in total all the physical interactions that can lead to self-reversal of magnetization have not been considered simultaneously in any of these works.

Hoffman [50] observed self-reversal on synthesized hemoilmenites with a composition that falls onto the center of the solid-solution series, thus indicating that the formation of cation-ordered regions in the crystals can occur at temperatures that are several hundred kelvins higher than the order–disorder transition temperature. But this is inconsistent with the model of self-reversal by Ishikawa and Syono. Therefore, for explaining the observed self-reversal effect, Hoffman suggested a refined model based on the Ishikawa–Syono concept. Upon cooling, the sample undergoes a lamination (phase separation) with the formation inside it of small regions rich in ilmenite, and regions rich in hematite. As cooling continues, the ilmenite regions ($T_{C,ilm} > T_{C,hem}$) become ordered first and serve as 'nuclei' for further ordering. The regions rich in hematite represent in this case the x phase. Upon contact of the hematite region with the surrounding region which became ordered upon contact with the ilmenite region, the hematite region becomes magnetized (after the T_C of hematite is passed) in the direction of the magnetizing field as a result of exchange interaction between this region (x phase) and the ordered phase. During further cooling, the ordered phase becomes magnetized antiparallel to the magnetizing field, which results in the formation of a reverse TRM. Hoffman [50] emphasizes that the exchange interaction between the x phase and the ordered phase forces their spins to align antiparallel, but the mechanism of spin ordering is not completely clear.

Later on, Hoffman refined this mechanism in paper [88], where it is again assumed that the x phase is nothing but an iron-rich cation-disordered boundary between two types of cation-ordered domains, and that the reverse TRM is only formed under the condition that both types of cation-ordered domains (ilmenite and hematite) exist in the grain in equal proportions. Hoffman [88] also assumes that the reverse TRM grows during sample cooling in a magnetic field via the rotation of isolated spins of iron inside cation-ordered domains.

In 1989, Nord and Lawson [89], when also refining the Ishikawa–Syono model, assumed that it is the double boundary around cation-ordered domains (WF phase) that plays the role of the x phase: upon cooling, it is the first to become magnetized (in the direction of the applied field), and the negative exchange interaction of the boundary with a cation-ordered domain (FiM phase) forces the cation-ordered region to become magnetized antiparallel to the applied field. This model was also used by Kennedy and Osborne [90] for explaining self-reversal of magnetization observed in dacitic samples.

In 1996, Hoffmann and Fehr [56] examined self-reversal of magnetization in the samples of dacitic pumice from the Pinatubo volcano (eruption of 1991). Based on micromagnetic and mineralogical investigations, they also ascribed the self-reversal effect to the exchange interaction between two interpenetrating hemoilmenite phases (WF and FiM). The WF phase is a surface phase having the form of a ring, and the FiM phase is the nucleus phase. The weak component (ring-shaped WF phase) acquires a TRM that is parallel to the applied field, whereas the ferrimagnetic (nucleus FiM) phase acquires a reverse TRM resulted from the negative exchange interaction of two hemoilmenite phases. The total TRM becomes reversed because of the higher saturation magnetization of the nucleus phase. According to the authors of Ref. [56], the transition zone between the two hemoilmenite phases can also play an important role in self-reversal.

After a magnetomineralogical investigation of 39 samples of dacitic pumices from the above-mentioned eruption of the Pinatubo volcano, Bina et al. [57] concluded that the self-reversal of magnetization observed in these samples can be explained using a model that is analogous to the Hoffmann–Fehr model [56], with the difference being that they suggested a zonal structure of the hemoilmenite phases instead of a rim–core ('shell') structure. The other reasoning remained unaltered.

It is worthy of note that the last two models do not differ fundamentally from the model suggested by Ishikawa and Syono in 1963 [37]: in all three models, the ordered FiM phase is magnetized antiparallel to the magnetizing field as a result of interaction with the iron-rich disordered phase, i.e., a WF phase or an intermediate x phase. The principal role here belongs to the coexistence of two phases (WF and FiM) in the same hemoilmenite crystal and their interaction leading to self-reversal of magnetization, rather than to the geometry and structure of these phases. This is confirmed by the results of Ozima et al. [37] who investigated the micromagnetic structure of hemoilmenite crystals of dacitic pumices from Haruna Mountain. The goal of the work [37] was to check the adequacy of the model suggested by Hoffmann and Fehr [56]. The micromagnetic investigations revealed the existence of two hemoilmenite phases but did not reveal either a skeleton [56] or a zonal [57] structure in the phases; nor has a boundary between the phases been found. Therefore, the authors of Ref. [37] concluded that neither the geometry nor the structure of the phases is essential for self-reversal. This is the only difference between the above two models and the Ishikawa–Syono model. Consequently, we may conclude that all three models represent, in principle, the same model of self-reversal.

In a number of works on self-reversal of magnetization in dacites from Haruna, Akagi, and Sambre mountains, Ozima et al. [37, 38, 91] came up with a proposal that the physical

mechanism responsible for self-reversal is also related to the exchange interaction at the boundary of two hemoilmenite phases. In Ref. [91], they studied dacitic pumices from the Akagi, On-take, and Sambre volcanoes, in which the carriers of the reverse TRM are hemoilmenite crystals with an ilmenite fraction of $0.4 \leqslant x \leqslant 0.7$. The results of these investigations were compared with those on self-reversal of magnetization, obtained in samples from mount Haruna [37] and volcano Pinatubo [53].

The micromagnetic observations performed using the Bitter powder technique revealed a multidomain structure of grains. For the most part, the grains were structureless, but a shell structure consisting of a ferrimagnetic core and an antiferromagnetic (plus weak ferromagnetism) rim was observed in the sample of dacitic pumice from the Sambre volcano. This structure of the grains strongly resembles the structure of grains in the samples from volcano Pinatubo [37, 56]. Hemoilmenite grains of the samples from the Akagi volcano possess a complex irregular structure with a strong component of the reverse TRM (sample designated as Akg in Ref. [37]); the sample with a weak TRM component (designated as Aka in Ref. [37]) has a clear ferrimagnetically ordered structure with parallel 180° domain walls. The irregular structure of hemoilmenite grains points to the fact that the sample was cooled so rapidly that the annealing time proved to be insufficient and, consequently, the ordered phase could not completely be formed in the hemoilmenite. The authors of Ref. [91] again emphasized that for a hemoilmenite crystal to acquire a reverse TRM it is necessary that two hemoilmenite phases coexist in the same grain, whereas the structure of the phases is of no decisive importance. Thus, the samples from the Haruna and Akagi volcanoes possess an irregular structure, whereas those from the Pinatubo and Sambre volcanoes exhibit a shell structure. However, the mechanism of self-reversal in all the samples was always the same.

According to Refs [46, 47, 87], the cooled weakly annealed hemoilmenite acquires an intense reversed TRM, whereas the strongly annealed hemoilmenite has a very weak reversed TRM. This conclusion is confirmed by the data obtained in the samples from the Akagi volcano. The authors of Ref. [91] concluded that the only presence of a certain chemical composition of the hemoilmenite (i.e., the amount of the ilmenite x-fraction), at which the hemoilmenite is capable of acquiring a reverse TRM, is insufficient for the self-reversal of the TRM: the process of the TRM self-reversal in hemoilmenites is equally affected by the rate of cooling of the sample, along with other thermal conditions of cooling. As to the samples of pumice taken from the On-take volcano, which do not exhibit properties of self-reversal, their hemoilmenite consists of a completely disordered WF phase.

Therefore, to show up the properties of self-reversal of magnetization it is insufficient to have samples that potentially can exhibit such properties. Of significant importance are also the conditions surrounding the experimental technique (cooling rate, time and temperature of annealing). All the ideas of the above-mentioned works by Ozima et al. [37, 38] had been formulated earlier; the authors simply confirmed them once again by experimental investigations of several series of samples. It is seen from the above that again everything reduces to the Ishikawa – Syono three-phase model of self-reversal, which, in turn, is based on Néel's idea concerning the self-reversal mechanism as a result of exchange interaction at the interphase boundary.

Let us consider in more detail the nanophase model of self-reversal of magnetization for the series of ilmenite – hematite solid solutions, suggested by Prévot et al. [58] in 2001, which is of interest, first and foremost, because the authors of the model believe that the model is applicable both to laboratory cases of self-reversal obtained in dacitic pumices of lavas from the Pinatubo volcano (eruption of 1991) and to samples taken from Haruna Mountain and dacitic andesites from the Nervado del Ruiz volcano (eruption of 1985). Thus, this phenomenological model might be considered universal to a certain extent. The main idea remains the same as in the models similar to the Ishikawa – Syono model: self-reversal of magnetization occurs due to the exchange interaction of two hemoilmenite phases with different degrees of cation ordering. A new point is the assumption that the ferrimagnetic cation-ordered domains spread in the cation-disordered antiferromagnetic matrix have a nanoscopic size.

In the above-described 'nanophase' model of self-reversal for a series of ilmenite – hematite solid solutions, the reversed (rTRM) and normal (nTRM) components of the TRM are carried by the cores and edges of partially cation-ordered nano-sized (observed in a microscope) domains. Because of the partial cation ordering, it is expected that both the core and the edge of each domain will behave ferrimagnetically at temperatures below corresponding Curie points. The idea of a 'shell structure' (core – edge) is borrowed from the model of self-reversal suggested by Hoffmann and Fehr [56].

Since, according to Ref. [58], it is most reasonable to suppose that each edge (x phase) will be slightly more enriched in Fe than the internal core, the edges are the first to acquire remanent magnetization (nTRM) upon cooling. Taking into account that the intralayer and interlayer superexchange interactions with nearest neighbors are ferromagnetic and antiferromagnetic, respectively, we can conclude that upon further cooling the net magnetic moment of the material of the core should become oppositely aligned (formation of an rTRM).

The nano-sized regions will in reality behave superparamagnetically in the absence of magnetic coupling with the adjacent substance; however, spins on the edges (x phase) should be coupled with the spins of the surrounding disordered matrix which should also be locally enriched in iron. If this is the case, the magnetization of the x phase can simultaneously be both highly coercive and thermostable, as indeed was established in experiment.

The above model is the last of the multiphase models of self-reversal of magnetization in hemoilmenites that have been suggested to date. Its detailed description shows that it contains nothing new as compared to the Ishikawa – Syono model [46, 47].

We will now consider mechanisms of self-reversal in continental and oceanic basalts containing titanomagnetites. As will be shown below, there are only scarce ideas here, as in the case of the self-reversal of magnetization of hemoilmenites. It is likely that this is again related to the above-mentioned 'narrowness of geography', i.e., with the small number of geographic points from which the samples have been taken.

In 1965, Havard and Lewis [59] supposed, based on the investigations of India basalts containing titanomagnetites, that the self-reversal of magnetization occurs as a result of a magnetostatic interaction between the titanomagnetite phase and the titanomaghemite phase formed as a result of a partial

oxidation of titanomagnetite (Néel two-phase mechanism). In 1975, Ryall and Ade-Hall [63] applied an analogous mechanism to explain the self-reversal obtained on samples of submarine basalts taken in the Central Atlantic; in 1979, Heller et al. [92] utilized this mechanism to explain the self-reversal of the NRM of continental basalts from Mount Etna (Sicily). A new statement by the authors of the last work is that the model of negative magnetostatic interaction requires a specific geometry: for the realization of self-reversal, some special geometrical conditions are required. Possibly, such conditions existed in the lava of 1669 that was studied by the authors. The authors of Ref. [92] supposed that this can be related to both the structure of the lava flux and the small rate of its cooling. Ryall and Hall [65] supported an analogous mechanism of self-reversal of magnetization observed in submarine basalts but added that the volume of titanomaghemite formed in the grain depended on the heating time. In Refs [66, 93], the above mechanism was used to explain the laboratory-observed self-reversal in both natural and synthesized titanomagnetites. The principal idea was by no means necessarily related to the formation of titanomaghemites upon oxidation, but rather simply to the presence of two phases of titanomagnetite with different degrees of oxidation in the same grain and to a magnetostatic interaction between them.

In 1966, O'Reilly and Banerjee [94] studied synthesized samples of titanomagnetites and, after obtaining self-reversal, supposed that it is also due to oxidation of titanomagnetite, which results in the formation of the Néel N type grains. This means that in fact we have here a single-phase mechanism of the N type but not in pure form, since chemical transformations (in particular, oxidation of titanomagnetite) play the principal role here.

In 1968, Ozima and Larson [61] studied submarine basalts from various regions of the Pacific Ocean and supposed that self-reversal of magnetization can be caused by the interaction of the titanomagnetite and titanomaghemite phases coexisting in a given magnetically ordered grain of the rock, but this interaction is of exchange character. The authors of Ref. [61] schematically explained the mechanism of self-reversal as follows. They introduced an A phase, which is the initial titanomagnetite phase with $T_C = 250\,^\circ$C, and a B phase, which is a secondary phase with $T_C = 300\,^\circ$C, the latter formed as a result of oxidation. If a sample contains two interpenetrating phases then, on cooling from $T = 300\,^\circ$C to T_0 in a magnetic field, the B phase becomes magnetized in the direction of the applied field immediately after the temperature falls to below $300\,^\circ$C. Approximately at $T = 250\,^\circ$C, the A phase will be magnetized under the influence of the B phase (negative interaction) antiparallel to the direction of the applied field. Conceivably, the total magnetization directly after the temperature is reduced to below $250\,^\circ$C might turn out to be parallel to the applied field and close to zero. On further cooling, the magnetization of the A phase will grow more rapidly than that of the B phase and at a certain temperature will be predominant. It is at this temperature that the self-reversal of magnetization will occur. It should be emphasized, following the authors of Ref. [61], that the sign of the reversed TRM critically depends on the relative content of the two phases, which, in turn, depends on the duration and temperature of heating.

Ozima and Larson [61] called attention to the fact that the mechanism of self-reversal of magnetization in ferrimagnets of deep-sea basalts differs from that observed in the samples from Haruna Mountain. The ferrimagnetic fractions of submarine basalts and samples from Haruna Mountain have different chemical compositions. In addition, the Haruna lavas undergo rapid cooling in nature, which reduces the possibility of high-temperature oxidation. In water, the titanomagnetite crystals can undergo low-temperature single-phase oxidation which leads to the formation of a titanomaghemite (at $T \sim 600\,^\circ$C).

In all the cases below where the titanomaghemite phase is mentioned it is supposed, unless otherwise stated, that it formed as a result of the oxidation of titanomagnetite, and this will no longer be emphasized separately. The idea of the self-reversal mechanism related to exchange interaction at the boundary between two phases coexisting in a single grain was used repeatedly for the explanation of laboratory-induced self-reversal (see, e.g., Refs [69, 95]).

In 1968 and 1976, Schult [62, 96] examined self-reversal on the samples of tertiary basalts also containing titanomagnetites and concluded that the self-reversal of magnetization occurs via a single-phase mechanism of the N type, but the grains become grains of the N type as a result of low-temperature oxidation of titanomagnetites. This idea was put forward earlier by O'Reilly and Banerjee [60], but Schult developed it further in more detail and studied the evolution of the type of grain spontaneous magnetization with progressing oxidation. Upon the oxidation of titanomagnetite, the $I_S(T)$ curve undergoes the following changes: $Q \to P \to L \to N$. Consequently, the titanomaghemites, in which self-reversal occurs due to ion redistribution, should have grains of the N type.

Schult also proposed for the first time, and Doubrovine and Tarduno [72] confirmed it by experimental data and developed it further, the following idea: apart from fine single-domain grains of the N type (with different degrees of oxidation, and different T_{comp} and T_C), the samples frequently contain multidomain weaker oxidized grains which can mask the signal from grains of the N type. Therefore, the $I_S(T)$ dependence in a sample with N type grains by no means always has a clearly pronounced TDM of the N type: in samples that exhibit two antiparallel components of the NRM, there is observed, instead of a clear minimum, either a wide spectrum of minima and compensation points at temperatures both above and below T_0 or 'humps' (then T_{comp} is not reached exactly). On samples with a single NRM component, a wide spectrum of minima and compensation points for $T < 0\,^\circ$C is observed. An analogous conclusion was reached by Trukhin et al. [1] and also on the basis of the model of self-reversal that occurs via the mechanism of the N type [97], which will be described in detail in Section 4.3.

Now, we consider in more detail the work by Doubrovine and Tarduno [72] in which self-reversal of magnetization was attained on titanomaghemites entering into oceanic basalts. According to this work, the mechanism responsible for the self-reversal is the single-phase mechanism of the Néel N type. The principal idea [72] is based on the approach (put forward in 1956 and 1962) by Verhoogen [23, 98], who was the first to suggest that it is ion redistribution in titanomaghemites that can be the cause of self-reversal of magnetization. The authors of Ref. [72] also apply the idea of Schult who, as was mentioned above, was the first to show that upon oxidation the titanomagnetite with $I_{SA} > I_{SB}$ is transformed into a titanomaghemite with $I_{SA} < I_{SB}$, i.e., grains of the N type are formed, and a self-reversal of the $I_S(T)$ occurs. The remanent magnetization of a sample with such pseudo-single-

domain grains will also suffer self-reversal. Based on the above-mentioned works, Doubrovine and Tarduno interpreted the experimentally exposed self-reversal effect as follows: the underwater low-temperature oxidation of basalts can lead to a redistribution of ions between the sublattices A and B, thus resulting in the formation of single-domain and pseudo-single-domain titanomaghemite grains of the N type (with $T_{\text{comp}} < T_0$), which are responsible for the observed partial self-reversal of the NRM and laboratory-induced partial and complete self-reversal of the PTRM and TRM.

A detailed investigation of the effect of self-reversal of magnetization on the samples of oceanic basalts containing titanomaghemites permitted the authors of the work [72] to hypothesize that a decrease (depending on the age) in the amplitudes of magnetic anomalies of the oceanic crust formed in the last 30 million years can occur not only as a result of the oxidation of the original magnetic minerals, but also as a result of the self-reversal of magnetization of minerals containing titanomaghemites.

It is pertinent to note that theoretical works on self-reversal of magnetization are quite rare. Among them, a series of works by Stephenson should be distinguished [99–102]. Let us consider them in more detail. In his first theoretical work (1969), Stephenson [99] showed that self-reversal can be related to cation redistribution which can take place upon oxidation. In 1972, Stephenson [100] calculated, in terms of a molecular-field approximation, the temperature dependences of spontaneous magnetizations of cation-deficient titanomagnetites and showed that with a certain cation distribution the TDM curves may be of the N type with a compensation point $T_{\text{comp}} > 450\,^{\circ}\text{C}$. In 1975, Stephenson [102] suggested the so-called model of magnetic shielding for the physical mechanism of self-reversal, according to which the phase with a greater T_{C} completely surrounded by a phase with a lower T_{C} may be magnetically shielded, as soon as its temperature, upon cooling, reaches the Curie temperature T_{C} of the surrounding low-temperature phase. Stephenson established that the shielding factor depends only on the relative magnitudes of magnetic permeabilities and demagnetizing factors of the two phases.

Nishida and Sasajima [103], who studied basalts from Göttingen (Germany) and Hiroshima (Japan), also revealed self-reversal. They undertook an attempt to establish whether the N type material is a cation-deficient titanomagnetite or a variety of the product that has formed as a result of cation redistribution. According to the authors of Ref. [103], the mechanism responsible for self-reversal is the single-phase Néel N type mechanism. These authors also studied submarine basalts containing titanomagnetites and synthesized titanomagnetites [64] to show that the grains of cation-disordered titanomagnetite (titanomaghemite) of the P or Q type can be transformed into N type grains on oxidation or cation ordering.

Let us now consider the mechanisms of self-reversal of chemical remanent magnetization (CRM) [78]. The CRM self-reversal was observed in hematite formed on heating maghemite. The experimental data obtained in Ref. [78] suggest that the self-reversal of remanent magnetization of hematite occurs only if the primary maghemite still remains blocked at the temperature of its transformation into hematite. When the transformation temperature is less than the blocking temperature T_{b} of maghemite and it is not blocked, the resulting remanent magnetization of hematite has a normal direction. McClelland and Goss [78] proposed a hypothesis that the self-reversal is a common feature of the magnetite – hematite transition and that a grain size plays an important role here. Since works devoted to self-reversal of magnetization caused by chemical transformations are very scarce, let us consider in more detail the process of self-reversal described in the work [78]. Originally, there exists a maghemite phase which is a carrier of the stable direct remanent magnetization. Then, there arise the first grains of hematite — superparamagnet — which is not a carrier of stable remanent magnetization until its volume is greater than the blocking volume at the reaction temperature. The process of increasing grain volume is equivalent to the process of decreasing temperature (the external temperature becomes less than the blocking temperature T_{b}). Thus, the authors of Ref. [78] suggest a system of 'cooling' in which hematite is a second phase for magnetic ordering. It is assumed that self-reversal of magnetization occurs as a result of exchange interaction at the interphase boundary between the maghemite and hematite, and that the greater the area of the boundary surface between the phases, the more clearly pronounced the self-reversal effect.

Above, we considered in some detail two-phase mechanisms of self-reversal of magnetization in titanomagnetites and hemoilmenites entering into rocks that have been studied by various authors, as well as the mechanism of self-reversal occuring as a result of chemical transformations. Now, we shall deal with the single-phase Néel N type mechanism of self-reversal and works in which this mechanism is applied to explaining the self-reversal of magnetization.

4.2 The Néel N type mechanism of self-reversal of magnetization

The numerous investigations of the phenomenon and physical mechanisms of self-reversal of magnetization of rocks, performed in our previous works [1, 2, 70, 80, 97], give grounds to advance the most likely physical mechanism of self-reversal which, in our opinion, is realized in most of the studied cases of self-reversal of magnetization in rocks.

When considering the problem of the most probable mechanism of self-reversal, we took into account that the principle of least action is operative in nature and that natural processes occur along the most rational paths. It was noted earlier that the most simple and elegant mechanism of self-reversal is Néel's mechanism caused by the change in the sign of the spontaneous magnetization I_{S} of the ferrimagnet crystal lattice with temperature or due to the diffusion of ions or electrons, although, possibly, by changes in some other thermodynamic parameters.

In Ref. [70], the authors for the first time concluded, based on experimental investigations of TRM self-reversal in Yakutia kimberlites, that the self-reversal in ferrimagnetic minerals of rocks occurs via the N type mechanism. Let us consider some results of these investigations. Figure 10 displays the complete self-reversal of the TM and TRM of a kimberlite sample from the Mir diamond pipe (Yakutia), which contained ferrimagnetic picroilmenites. Curve *1* shows the temperature dependence of the initial magnetic susceptibility $\varkappa_0$ which is inversely proportional to the coercive force ($H_{\text{C}} \sim A/\varkappa_0$, where A is some constant). The change in the sign of the TM (curve *2*) occurs at a temperature T at which $\varkappa_0$ reaches a maximum, while H_{C}, correspondingly, reaches a minimum, as is predicted by the N type mechanism. Curve *3* corresponding to the development of TRM upon cooling the

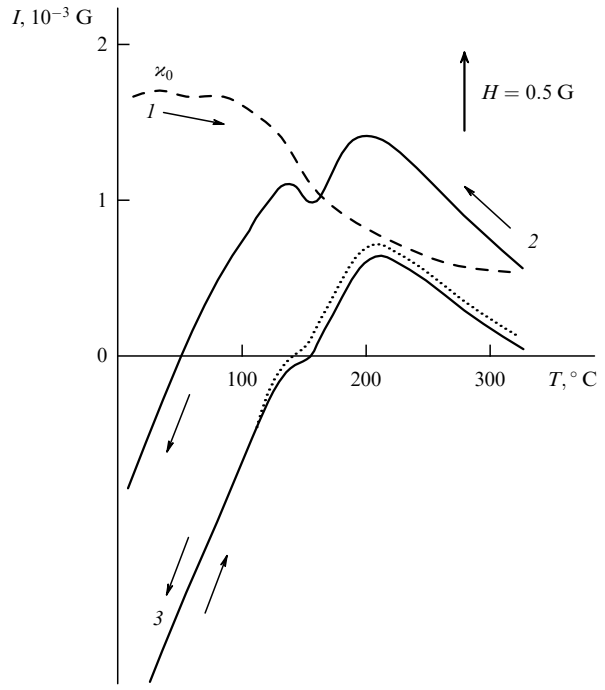

Figure 10. Self-reversal of total thermo magnetization and thermo remanent magnetization of a kimberlite sample (Mir diamond pipe, Yakutia) containing ferrimagnetic picroilmenites (natural analogs of hemoilmenites) upon cooling from $T = 350\,^{\circ}$C to $T_0 \approx 20\,^{\circ}$C in a field of $H = 0.5$ G: (*1*) variation of the initial magnetic susceptibility upon heating the sample in a field H; (*2*) formation of thermomagnetization upon cooling the sample in a field of $H = 0.5$ G, and (*3*) temperature dependence of thermoremanent magnetization $I_{rT}(T)$, obtained upon heating of the sample; the dotted line corresponds to the temperature dependence of the thermoremanent magnetization $I_{rT}(T)$, obtained upon cooling of the sample.

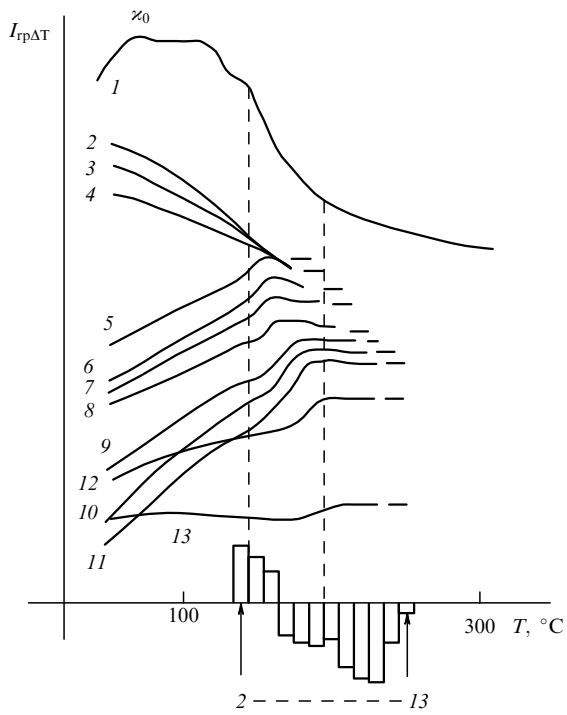

Figure 11. Spectra of partial remanent magnetizations $I_{rp\Delta T}$ arising on intervals $\Delta T = 10\,^{\circ}$ and $H = 1$ G, and their temperature dependences for a kimberlite sample (Mir diamond pipe, Yakutia) containing ferrimagnetic picroilmenites (natural analogs of hemoilmenites). In each measurement (curves *1–13*), the field H was switched on only in the specified temperature interval ΔT, and the preceding and subsequent coolings were performed at $H = 0$; the magnitude of $I_{rp\Delta T}$ was measured after the temperature T reached T_0. Some $I_{rp\Delta T}(T)$ curves are vertically shifted for convenience with respect to one another; the axis of zero magnetization is marked by dashes located to the right of the origin of each curve.

sample in a magnetic field of $H = 0.5$ G passes through zero at a temperature that is somewhat higher (by 100 °C) than the curve of the TM formation. This is easily comprehended, since the TM additionally includes the magnetization $\varkappa_0 H$ which is always directed along the field **H**.

Some samples taken from the Mir diamond pipe exhibited an anomalous behavior of the NRM. Figure 7 displays cyclic thermodemagnetization of the NRM in the absence of a field ($H = 0$). It is seen that on heating (curve *1*) the NRM increases rather than decreases as should be expected from the viewpoint of the physics of magnetic phenomena. Curves *3* and *4* pass into the negative half-plane. It is clear that such a behavior of the NRM is related to the complex character of sample magnetization in nature over many millions of years.

Figure 11 depicts the $\varkappa_0(T)$ dependence (curve *1*) and the temperature dependences of PTRM upon cooling of the sample in a zero field (curves *2–13*); also given are the magnitudes and directions of the PTRMs obtained in various temperature ranges upon cooling in a zero field (the measurements were carried out at $T = 20\,^{\circ}$C).

Note that the transition from negative to positive PRTMs occurs at temperatures at which a sharp increase in $\varkappa_0$ (and, consequently, a decrease in H_C) is observed.

Figure 12 shows demagnetization curves for positive and negative PTRMs in an alternating magnetic field h with a frequency of 50 Hz. It is seen that all of them are single-phase. If the magnetization emerged in a two-phase mineral system, the $I_{rp\Delta T}$ (h) curves of the demagnetization of the positive and

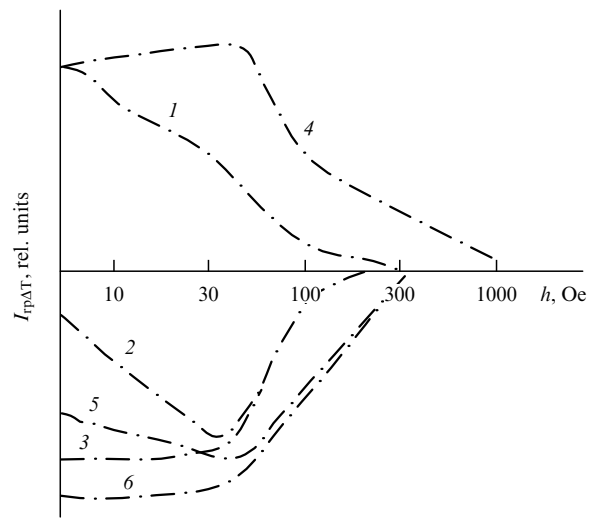

Figure 12. Demagnetization (in an alternating magnetic field) of two kimberlite samples (Mir diamond pipe, Yakutia) containing ferrimagnetic picroilmenites, which have different partial remanent magnetizations $I_{rp\Delta T}$: (*1*)–(*3*) a sample with the coercive force $H_C = 95$ Oe; (*4*)–(*6*) a sample with $H_C = 280$ Oe; (*1*) $I_{rp\Delta T}$ demagnetization was produced by switching on a field of $H = 200$ Oe on a temperature interval ΔT from 300 to 20 °C; (*2*) a field of $H = 1$ Oe in the same temperature interval; (*3*) a field of $H = 200$ Oe in an interval $\Delta T = 300 - 220\,^{\circ}$C; (*4*) $H = 200$ Oe, $\Delta T = 300 - 20\,^{\circ}$C; (*5*) $H = 4$ Oe, $\Delta T = 300-20\,^{\circ}$C, and (*6*) $H = 4$ Oe, $\Delta T = 300-240\,^{\circ}$C.

Table. Conditions of synthesis of hemoilmenites [1]

Sample No.	Starting products	$\chi_{\rm ilm}^{\rm non}$	τ, day	p, kbar	T, °C	Buffer	Final product
970	Hem + TiO_2	0.62	3	5	900	> NNO	ilm
971	Hem + TiO_2	0.55	3	5	900	> NNO	ilm
981	Hem + TiO_2	0.80	7	5	750	NNO	ilm
987	Hem + TiO_2	0.52	3	5	790	HM	ilm + Rt
1007	Mag + Rt + *	0.77	4	5	840	NNO	ilm
1024	Mag + Rt + *	0.70	4	5	870	> NNO	ilm
997	Mag + Rt	0.90	11	5	790	QFM	ilm + Rt
1000	Hem + TiO_2 + *	0.52	9	5	790	HM	ilm + Rt
1005	Hem + TiO_2 + *	0.48	12	5	790	HM	ilm + Rt
1019	Ilm + Rt + Psb	0.43	0.83	5	900	HM	ilm + Psb
1045	Mag + Rt	0.52	12	5	790	HM	ilm + Rt
1043	Hem + TiO_2	0.52	6	5	790	HM	ilm + Rt

Designations: $\chi_{\rm ilm}^{\rm non}$ is the gross composition of the initial mixture; τ, the time of synthesis; p, the pressure during synthesis; T, the synthesis temperature; ilm, ilmenite; Hem, hematite; Mag, magnetite; Rt, rutile; Psb, pseudobrookite; *, 5% $^{57}Fe_2O_3$; HM, hematite – magnetite buffer; NNO, Ni – NiO buffer; QFM, quartz – fayalite – magnetite buffer; > NNO, the fugacity of oxygen is greater than for the NNO buffer.

negative PTRMs in an alternating magnetic field h would also be two-phase.

The represented experimental dependences are already sufficient to state that the self-reversal of magnetization is realized in a single mineral phase and, consequently, it occurs via the Néel N type mechanism.

However, in natural samples of rocks two- and multiphase mineralogical systems can always be presented. Therefore, using electron-microprobe analysis, we determined the composition of the main fraction of the self-reversing samples. In the hemoilmenites $x{\rm FeTiO_3}(1-x){\rm Fe_2O_3}$ studied in Ref. [70], $x = 0.5-0.7$. Proceeding from these data, synthesized hemoilmenites of composition $x \approx 0.5-0.9$ have been prepared at the Institute of Experimental Mineralogy, Russian Academy of Sciences (Chernogolovka, Moscow region) with whose researchers we carried out cooperative work on the investigation of the composition and magnetic properties of synthesized hemoilmenites.

The synthesized hemoilmenites (the conditions of their synthesis are given in the table) differed in x [104]. The most clearly pronounced self-reversal effect was noted in samples of single-phase hemoilmenites with $x = 0.62$ (sample No. 970, see the table) and $x = 0.55$ (sample No. 971, see the table and Fig. 6) and on some other samples of a similar composition [1].

The results of this experiment, in our opinion, provide a decisive argument in favor of the Néel N type mechanism of self-reversal.

Consider now, in connection with the self-reversal of TRM, the theory of the N type mechanism and N type $I_{\rm S}(T)$ curves (see Section 3.1). Assume that $\lambda < \mu$. According to Eqn (3.4), we arrive at the relationships

$$|I_{\rm SB}| > |I_{\rm SA}| \quad \text{and} \quad I_{\rm S} = I_{\rm SA} - I_{\rm SB} < 0\,. \tag{4.1}$$

For the case where the $I_{\rm S}$ sign is changed with increasing temperature, the following relationships should be fulfilled near the Curie point (at constant λ and μ):

$$|I_{\rm SB}| < |I_{\rm SA}|\,, \qquad I_{\rm S} = I_{\rm SA} - I_{\rm SB} > 0\,. \tag{4.2}$$

According to Ref. [22], the expression for the boundary separating ferrimagnetic phases with an opposite direction of the spontaneous magnetization $I_{\rm S}$ is written as follows:

$$\lambda(\alpha+1) - \mu(\beta+1) = 0\,. \tag{4.3}$$

The $I_{\rm S}(T)$ curves exhibiting a compensation point (N, V types) are obtained in the range of parameters α and β that lies below the boundary described by Eqn (4.3), i.e., when the condition

$$\frac{\beta+1}{\alpha+1} > \frac{\lambda}{\mu} \tag{4.4}$$

is valid.

Thus, the fulfillment of inequality (4.4) is a condition for the change in the sign of $I_{\rm S}$ with increasing temperature from 0 K to $T_{\rm C}$.

Referring to paper [22], the $I_{\rm S}(T)$ curves of type N should possess a compensation point. However, $T_{\rm comp}$ will only be examined experimentally in the case of a homogeneous uniform ferrimagnet possessing a single Curie point. In reality, a real ferrimagnet exhibits a diffuse ferrimagnet – paramagnet phase transition at $T_{\rm C} \pm \Delta T_{\rm C}$, which leads to a corresponding smear of the compensation point $T_{\rm comp}$. Therefore, the $I_{\rm S}(T)$ curve in sufficiently strong fields H will not reach zero; only a slight decrease in $I_{\rm S}$ in the range of $T_{\rm comp} \pm \Delta T_{\rm comp}$ will occur.

The investigated samples of hemoilmenite consist of separate grains which can have slightly different $T_{\rm C}$ and $T_{\rm comp}$. As a result, the $I_{\rm S}(T)$ and $I_{\rm rS}(T)$ curves characterizing the grain ensemble will possess either a concave shape (Fig. 13a) or 'tails' (Fig. 13b) [1]. Thus, we suppose that the concave shape of $I_{\rm rS}(T)$ curves that was observed in self-reversing hemoilmenites is related to the presence of a spectrum of compensation points and blocking temperatures $T_{\rm b}$, and, correspondingly, Curie points $T_{\rm C}$.

It is known that in a saturation field ($H = H_{\rm S}$) under cooling of homogeneous ferrimagnets in a magnetic field that is greater or equal to the saturation value ($H \geqslant H_{\rm S}$) a compensation point $T_{\rm comp}$ is observed in the experimental $I_{\rm S}(T)$ curves, but no self-reversal of $I_{\rm S}$ occurs, since the strong external field for $T < T_{\rm comp}$ turns around the magnetic moments that changed their signs again along the field. For the self-reversal of magnetization to occur, it is necessary that the relationship

$$H < H_{\rm C}(T) \tag{4.5}$$

be fulfilled for all $T \leqslant T_{\rm comp}$. Therefore, as a rule, the self-reversal of TRM is observed in relatively weak fields (see, e.g., Refs [27, 70]).

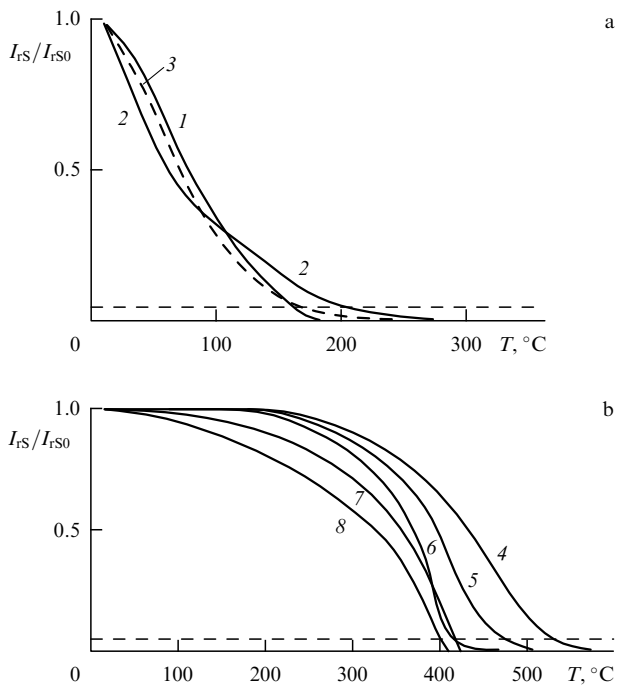

Figure 13. Curves of the temperature dependence of remanent saturation magnetization $I_{rS}(T)/I_{rS0}$ of the samples of synthesized hemoilmenites $x\mathrm{FeTiO_3}(1-x)\mathrm{Fe_2O_3}$ with a molecular fraction x of ilmenite (see the table) (the curves were obtained on heating the samples in a field of $H = 0$): (*1*) sample No. 970, $x = 0.62$; (*2*) sample No. 971, $x = 0.55$; (*3*) sample No. 981, $x = 0.80$; (*4*) sample No. 997, $x = 0.90$; (*5*) sample No. 1005, $x = 0.48$; (*6*) sample No. 1000, $x = 0.52$; (*7*) sample No. 1019, $x = 0.43$, and (*8*) sample No. 1045, $x = 0.52$.

Thus, we suppose that the following physical model of self-reversal of magnetization in the hemoilmenites studied is realized. In each ith grain for $T_{\mathrm{comp}\,i} < T < T_{\mathrm{C}i}$ in a field H, the thermomagnetization occurs along the direction $I_{\mathrm{SA}} > I_{\mathrm{SB}}$. For temperatures $T < T_{\mathrm{comp}\,i}$, the magnetization I_{SB} of the sublattice B starts exceeding I_{SA}. If the applied field H is less than $H_{\mathrm{C}i}(T)$ for $T < T_{\mathrm{comp}\,i}$, then there occurs an increase in the negative I_{T} as T decreases. After the field H is switched off, the reversible part of I_{T}, directed along the field, disappears; as a result, the absolute value of $I_{\mathrm{rT}}(T_0)$ becomes more than that of $I_{\mathrm{T}}(T_0)$. The superposition of the $I_{\mathrm{T}\,i}(T)$ and $I_{\mathrm{rT}\,i}(T)$ curves of the entire ensemble of hemoilmenite grains leads to the experimentally examined regularities in the evolution of the TRM self-reversal.

In 2005, Bezaeva et al. [97] developed a computerized model of the self-reversal phenomenon based on the single-phase Néel N type mechanism; in terms of this model, various types of curves of the temperature dependence of magnetization with self-reversal have been obtained. The $I_{\mathrm{T}}(T)$ dependences calculated in the framework of this computerized model coincide qualitatively with analogous experimental dependences [1, 2, 70].

4.3 Computer model of the phenomenon of self-reversal of magnetization based on the Néel N type physical mechanism

In attempting to explain self-reversals of magnetization obtained under laboratory conditions by the realization of the Néel N type mechanism, it is assumed that the rock exhibiting the property of self-reversal contains ferrimagnetic grains of the Néel N type, i.e., those in which the spontaneous magnetization changes sign with temperature (see Fig. 5). As was shown in Section 3, this is possible only in a narrow range of magnetic parameters, in particular, of molecular-field constants α and β that determine the type of the temperature dependence of spontaneous magnetization (N, P, etc.; see Fig. 5).

Let us consider an ensemble of uniaxial single-domain noninteracting ferrimagnetic grains of the N type and the processes of magnetization of such an ensemble in a constant external magnetic field H. Such a consideration simulates a standard laboratory experiment on thermomagnetization (or on thermodemagnetization) of a sample of rock; the rock sample is replaced with the above-described ensemble, and the process of thermomagnetization with a point-by-point calculation of the corresponding curve of the temperature-dependent magnetization (TDM).

In a rock, the ferrimagnetic grains occupy only a few percent or even fractions of a percent of the entire volume; therefore, the interaction of the ferrimagnetic grains with each other is negligibly small as compared to the interaction of each grain with the magnetizing field. The grains are assumed to be single-domain, since rocks with such grains are quite frequently encountered in nature and are the main carriers of paleomagnetic information.

Let us briefly consider the main principles of simulation. It is convenient for us to employ all the equations in a dimensionless form; therefore, the magnetizations of the grain ensemble and of a single grain are divided by I_{S0} (spontaneous magnetization at $T = 0$ K) and, consequently, are dimensionless. Since the grains are assumed to be noninteracting, the total magnetization of the ensemble of n grains at a temperature point τ (where $\tau \in [0, 1]$ is the reduced temperature: $\tau = T/T_{\mathrm{C}}$) is determined as a vector sum of corresponding magnetizations of individual grains:

$$\frac{\mathbf{I}(\tau)}{I_{\mathrm{S0}}} = \sum_{i=1}^{n} \frac{\mathbf{I}(\tau)}{I_{\mathrm{S0}}}\,, \tag{4.6}$$

while the magnetization $\mathbf{I}_i(\tau)$ of an individual grain is obtained as the projection of the spontaneous magnetization $\mathbf{I}_{\mathrm{S}i}(\tau)$ of the ith grain onto the direction of the applied magnetic field H (see Fig. 14):

$$\frac{\mathbf{I}_i(\tau)}{I_{\mathrm{S0}}} = \sum_{i=1}^{n} \frac{\mathbf{I}_{\mathrm{S}i}(\tau)}{I_{\mathrm{S0}}} \cos\left(\theta_i(\tau)\right), \tag{4.7}$$

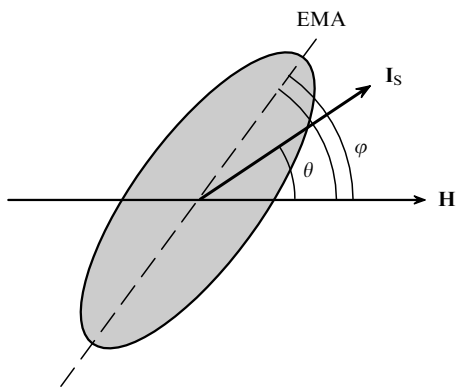

Figure 14. Schematic of a uniaxial single-domain ferrimagnetic grain in a magnetic field H: I_{S} is the spontaneous magnetization of the ferrimagnet composing the grain; EMA is the easy magnetic axis; φ is the angle between the EMA and the vector of strength $\mathbf{H}$ of the applied magnetic field, and θ is the angle between the vectors $\mathbf{I}_{\mathrm{S}}$ and $\mathbf{H}$, corresponding to a minimum of the system's energy.

where $\mathbf{I}_{Si}(\tau)$ is determined according to the Néel theory of collinear ferrimagnetism [see Eqns (3.4, (3.5)]. In further calculations, the above-mentioned equations will be used in the reduced form. To this end, we introduce the following designations:

$$\widetilde{\mathbf{I}}_{\mathrm{SA}} = \frac{\mathbf{I}_{\mathrm{SA}}}{\lambda I_{\mathrm{S0}}}\,, \quad \widetilde{\mathbf{I}}_{\mathrm{SB}} = \frac{\mathbf{I}_{\mathrm{SB}}}{\mu I_{\mathrm{S0}}}\,, \quad \widetilde{\mathbf{I}}_{\mathrm{S}} = \frac{\mathbf{I}_{\mathrm{S}}}{I_{\mathrm{S0}}}\,, \quad \delta = \frac{J+1}{3J}\,. \tag{4.8}$$

With allowance for these designations, Eqns (3.4) and (3.5) take on the form

$$\widetilde{\mathbf{I}}_{\mathrm{SA}} = B_J\left(\frac{1}{\tau}\left(\frac{\alpha\lambda}{\delta\mu}\,\widetilde{\mathbf{I}}_{\mathrm{SA}} - \frac{1}{\delta}\,\widetilde{\mathbf{I}}_{\mathrm{SB}}\right)\right), \tag{4.9}$$

$$\widetilde{\mathbf{I}}_{\mathrm{SB}} = B_J\left(\frac{1}{\tau}\left(\frac{\beta}{\delta}\,\widetilde{\mathbf{I}}_{\mathrm{SB}} - \frac{\lambda}{\delta\mu}\,\widetilde{\mathbf{I}}_{\mathrm{SA}}\right)\right),$$

$$\widetilde{\mathbf{I}}_{\mathrm{S}} = \lambda\widetilde{\mathbf{I}}_{\mathrm{SA}} + \mu\widetilde{\mathbf{I}}_{\mathrm{SB}}\,. \tag{4.10}$$

It should be specially emphasized that Eqns (4.9) and (4.10) define both the magnitudes and the signs of $\widetilde{\mathbf{I}}_{\mathrm{SA}i}(\tau)$, $\widetilde{\mathbf{I}}_{\mathrm{SB}i}(\tau)$, and $\widetilde{\mathbf{I}}_{\mathrm{S}i}(\tau)$ in the case of an ensemble of identical grains with $\widetilde{\mathbf{I}}_{\mathrm{S}i}(\tau) = \widetilde{\mathbf{I}}_{\mathrm{S}}(\tau)$.

The angle $\theta_i(\tau)$ in Eqn (4.7) is the angle between $\mathbf{I}_{\mathrm{S}}(\tau)$ and $\mathbf{H}$ (see Fig. 14) at a certain value of which the total energy of a uniaxial single-domain ferrimagnetic grain in an external magnetic field becomes a minimum. The total energy represents a sum of the energy of uniaxial form anisotropy, the energy of magnetocrystalline anisotropy, and the energy of the magnetic moment in the external magnetic field. The density of the total energy of such a system is expressed by the following equation (in a dimensionless form):

$$\begin{aligned}\widetilde{E}(\tau) &= \frac{E(\tau)}{E_0} = \frac{E_{\mathrm{A}}^{\mathrm{F}}(\tau)}{E_0} + \frac{E_{\mathrm{A}}^{\mathrm{CR}}(\tau)}{E_0} + \frac{E_H(\tau)}{E_0} \\ &= -\frac{\pi I_{\mathrm{S0}}}{H_0}\left(\frac{I_{\mathrm{S}}(\tau)}{I_{\mathrm{S0}}}\right)^2 \cos^2\left(\varphi - \theta(\tau)\right) \\ &\quad - \frac{|K_{10}|}{I_{\mathrm{S0}}H_0}\left(\left|I_{\mathrm{SA}}^3(\tau)\right| + \left|I_{\mathrm{SB}}^3(\tau)\right|\right)\cos^2\left(\varphi - \theta(\tau)\right) \\ &\quad - \frac{|\mathbf{I}_{\mathrm{S}}(\tau)|}{I_{\mathrm{S0}}}\,\frac{|\mathbf{H}|}{H_0}\cos\left(\theta(\tau)\right),\end{aligned} \tag{4.11}$$

where $E_{\mathrm{A}}^{\mathrm{F}}$, $E_{\mathrm{A}}^{\mathrm{CR}}$, and E_H are the energy densities of the uniaxial form anisotropy, magnetocrystalline anisotropy, and magnetic moment in a magnetic field H, respectively; $E_0 = I_{\mathrm{S0}}H_0$, and $H_0 = 1$ G; φ is the angle between the vector $\mathbf{H}$ and the easy magnetic axis (EMA), and K_{10} is the first anisotropy constant at $T = 0$ K. In what follows, the energy densities of the form anisotropy, magnetocrystalline anisotropy, and total anisotropy (magnetocrystalline anisotropy and form anisotropy) will be denoted as E^{F}, E^{CR}, and E_{A}, respectively.

The angle $\theta_i(\tau)$ is determined by simultaneously solving Eqn (4.11) and the following set of equations:

$$\begin{aligned}&\frac{\partial\widetilde{E}(\tau)}{\partial\theta} = 0\,, \\ &\frac{\partial^2\widetilde{E}(\tau)}{\partial\theta^2} > 0\,.\end{aligned} \tag{4.12}$$

Each concrete value of $\theta_i(\tau)$ depends on the relation between $|E_{\mathrm{A}}|$ and $|E_{\mathrm{H}}|$ or, equivalently, between the strength H of the magnetizing field and the coercive force H_{C} of a grain, which is an energy barrier hindering magnetization. The magnetization of a ferrimagnetic grain is considered as a turn of the vector of spontaneous magnetization from the position along the EMA toward the field direction (see Fig. 14).

The theoretical TDMs of the ensemble of ferrimagnetic grains were calculated from Eqns (4.6)–(4.12). In computations, the angles φ_i, $i = \overline{1, n}$, where n is the number of grains in the ensemble, were set from the values lying in the interval [0, $\pi/2$].

The calculated TDMs correspond to experimental $I_{\mathrm{T}}(T)$ dependences. Rocks contain, as a rule, ferrimagnetic grains of various chemical compositions, which, consequently, are characterized by different molecular-field constants α and β. This was also taken into account in calculations.

4.3.1 Results of computer simulation. Now, we turn to the results of computer simulation and first consider the case of an ensemble of identical ferrimagnetic grains. All the equations used in simulation were reduced to a dimensionless form; the strength of the magnetic field was normalized and, therefore, was also dimensionless. In this connection, we introduce concepts of a 'weak field' and 'strong field'. By the weak field H_{w}, we mean a field whose strength is less than the coercive force of the ensemble of grains in the entire temperature range under consideration or a field at which $|E_{\mathrm{A}}| > |E_H|$ (also at all temperatures); by the strong field, we mean a field H_{st} for which the opposite inequalities are valid, i.e., $H > H_{\mathrm{C}}$ and $|E_{\mathrm{A}}| < |E_H|$. All the other fields are assumed to be 'intermediate'.

In a weak field H_{w} (Fig. 15, curve *1*), the simulation yields a TDM of type 1, which is characterized by a single compensation point (CP) and by the presence of a self-reversal effect. In a strong field H_{st}, the self-reversal effect disappears but the CP is retained (see Fig. 15, curve *2*); this corresponds to a TDM of type 2 which, along with the TDM of type 1, forms two classical limiting cases. However, in the model under consideration, a 'transient' TDM or a TDM of

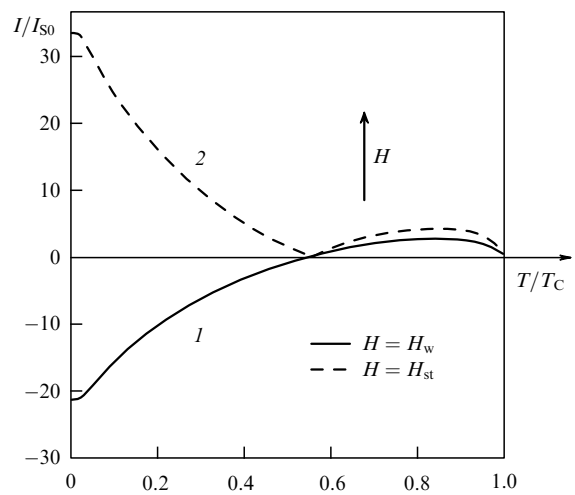

Figure 15. Temperature dependences of magnetization of type 1 (curve *1*) and type 2 (curve *2*) for an ensemble of identical uniaxial single-domain ferrimagnetic grains in a 'weak' magnetic field H_{w} and in a 'strong' magnetic field H_{st}, respectively.

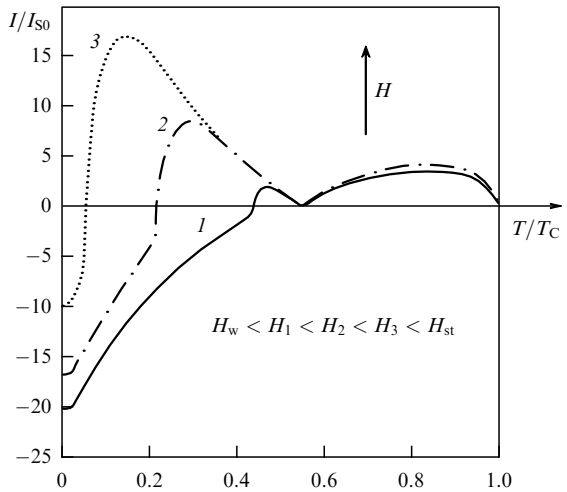

Figure 16. Temperature dependences of magnetization of type 3a for an ensemble of identical uniaxial single-domain ferrimagnetic grains obtained at various values of the strength of the external magnetic field: $H_w < H_1 < H_2 < H_3 < H_{st}$.

type 3 is realized in the case of $H_w < H < H_{st}$, which is characterized by two compensation points and by the presence of a self-reversal effect (Fig. 16).

The concrete shape of a TDM of type 3 depends on the relationship between the magnitudes of the energy density $|E^F|$ of form anisotropy and the energy density $|E^{CR}|$ of magnetocrystalline anisotropy, whereas in Eqn (4.11) this relationship, in turn, depends on concrete constants of the substance and, in particular, on the values of the spontaneous magnetization I_{S0} and the first anisotropy constant K_{10} at $T = 0$ K. These constants for various ferrimagnets vary within sufficiently wide ranges: 10 G $\leqslant I_{S0} \leqslant 10^3$ G, and 10^4 erg cm^{-3} $\leqslant K_{10} \leqslant 10^6$ erg cm^{-3}. At a proper choice of constants, a case can be realized where the energy of uniaxial form anisotropy far exceeds the energy of magnetocrystalline anisotropy: $|E^F| \gg |E^{CR}|$. In these conditions, the simulation yields a TDM of type 3a (see Fig. 16). In the opposite case ($|E^F| \ll |E^{CR}|$), we arrive at a TDM of type 3b (Fig. 17).

Notice that the two CPs in the TDM curve of type 3a have a fundamentally different nature: the higher-temperature CP is related to the change in the sign of I_S of a ferrimagnet of type N, whereas the presence of the second CP is due to processes of magnetization of an ensemble of grains. The temperature dependence of magnetization of type 3a is called 'transient', since for $H > H_w$ the lower-temperature CP in the TDM curve of type 3a is shifted with increasing H to the left along the temperature axis until H becomes equal to H_{st} and the TDM curve passes completely to the positive half-plane, i.e., until the TDM of type 3a transforms into a TDM of type 2.

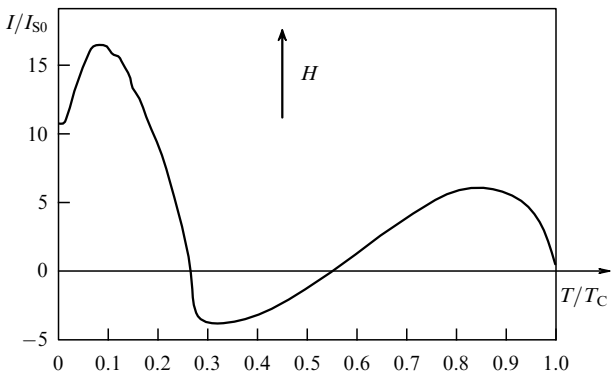

Figure 17. Temperature dependence of magnetization of type 3b for an ensemble of identical uniaxial single-domain ferrimagnetic grains in a magnetic field H ($H_w < H < H_{st}$).

Next, we consider an ensemble of type-N ferrimagnetic grains which have different magnitudes of the molecular-field constants α and β. The temperature dependence of the magnetization of type 1 does not differ from that of the ensemble of identical grains, except for the fact that the CP is now a certain average CP rather than referring to a concrete grain from the ensemble. Contrary to the TDM of type 1, the TDM of type 2 changes its shape: the CP disappears, which is related to the effect of averaging (Fig. 18a). In some works, their authors performed the tests for revealing grains of the N type, trying to obtain a TDM of type 2 in a pure form on samples of rocks possessing self-reversing properties. When no CP is revealed (as in Fig. 18a), they concluded that no grains of N type were present in the sample studied and excluded the possibility of the realization of the single-phase Néel N type mechanism as responsible for the observed self-reversal of the TRM, which may be erroneous. It is important to note that the TDM of type 2 for the ensemble of nonidentical grains has a concave shape.

Trukhin et al. [1] noted that all (without exception) curves of thermodemagnetization of NRM, obtained in samples of synthesized hemoilmenites which subsequently exhibited self-reversal of the TRM upon thermomagnetization, also possessed a concave shape. The authors concluded that it is precisely the concave shape of the curve of thermodemagnetization of the NRM rather than the presence of a CP that directly indicates that the samples offer self-reversing properties. This conclusion is confirmed by the results of simulation.

The temperature dependence of magnetization of type 3 (3a and 3b) for ferrimagnetic grains with different α and β can exhibit both two CPs and, as a result of averaging, a single CP (Fig. 18b).

4.3.2 Comparison of the results of modeling with experimental data. Let us compare some results of simulation with available experimental data. Figure 19 depicts curves of thermodemagnetization of the TRM of samples containing natural analogs of hemoilmenites [70]. It is seen that with increasing the field strength the compensation point is shifted to the left along the temperature axis until the TDM in a field of 4 G completely passes to the positive half-plane. Similar results were obtained upon simulation. The most clearly pronounced TDM of type 3a was obtained on the same sample [70] upon its thermomagnetization in a field of 0.5 G (see Fig. 10). Although the second CP is not reached, the tendency is clearly observed. In terms of the model suggested, this can be explained by the averaging effect because of the presence of ferrimagnetic inclusions of various kinds in the sample.

As was described in Section 4.2, the authors of Ref. [70] proposed a hypothesis that the observed self-reversal of magnetization occurs via the Néel N type mechanism; to confirm this, they synthesized hemoilmenites with a composition similar to that of natural hemoilmenites [1]. And, again, self-reversal of magnetization was obtained (see Fig. 6), which

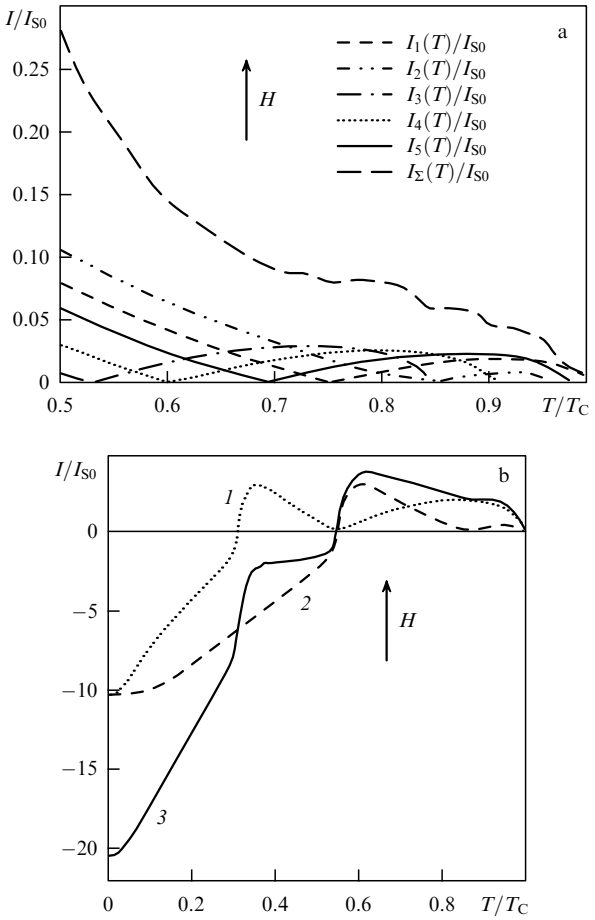

Figure 18. (a) The upper curve corresponds to the temperature dependence of magnetization of type 2 for an ensemble consisting of $n = \sum_{i=1}^{5} n_i$ uniaxial single-domain ferrimagnetic grains of various chemical composition (characterized by different molecular-field constants α_i and β_i, Curie points T_{Ci}, and compensation points T_{compi}). This curve represents a superposition of all the lower curves, each corresponding to a temperature-dependent magnetization (TDM) of an ensemble of n_i identical ferrimagnetic grains. T_C stands for the maximum (among all T_{Ci}) Curie point. (b) Temperature dependences of magnetization of type 3a for an ensemble of n_1 identical uniaxial single-domain ferrimagnetic grains characterized by molecular-field constants α_1 and β_1 (curve *1*) in a field H ($H_w < H < H_{st}$) and of an ensemble of n_2 uniaxial single-domain ferrimagnetic grains in a field H (characterized by molecular-field constants α_2 and β_2) (curve *2*); curve *3* is a superposition of curves *1* and *2* relevant to the TDM of type 3a for an ensemble of $n = n_1 + n_2$ nonidentical uniaxial single-domain ferrimagnetic grains in a field H (of which n_1 are characterized by molecular-field constants α_1 and β_1 and the other n_2 grains by the constants α_2 and β_2).

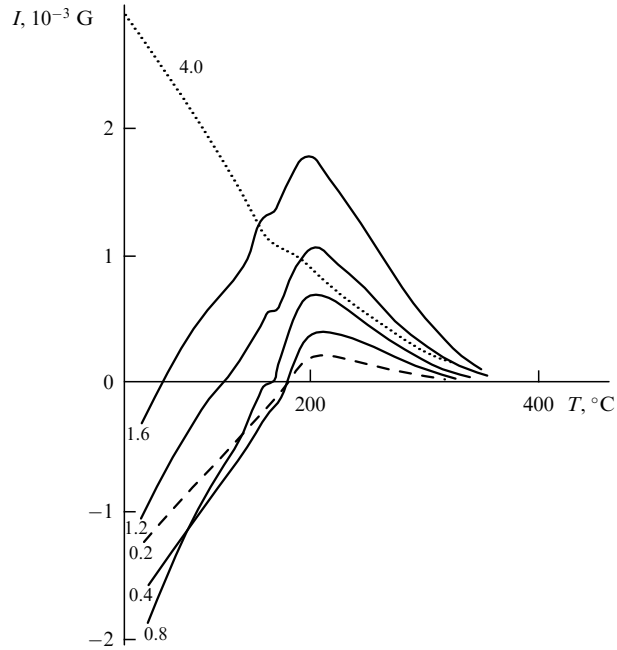

Figure 19. Dynamics of the suppression of self-reversal of magnetization in a kimberlite sample (Mir diamond pipe, Yakutia, Russia) containing ferrimagnetic picroilmenites (natural analogs of hemoilmenites) with increasing magnetic-field strength H from 0.2 to 4.0 G.

proves the hypothesis submitted. All the thermodemagnetization and thermomagnetization curves plotted in Fig. 6 (exclusive of the first one) are in keeping with TDMs of type 1.

Now, we turn to the work [2]. The experimental curves of the TDM obtained on samples of oceanic basalts containing natural titanomagnetites demonstrate self-reversal, namely, a classical TDM curve of type 1. However, in Ref. [80], where self-reversal of magnetization was obtained on a modified Berlin blue (Fig. 20a), the TDM curve corresponded to type 3b. The self-reversal that was obtained in the same work on a sample of nickel salt of formic acid is illustrated in Fig. 20b. TDM curve *1* corresponds to cooling in a magnetic field and is a TDM curve of type 1, whereas curve *2* corresponds to heating of the sample that was preliminarily cooled in the absence of a magnetic field and, in fact, also is a TDM curve of type 1 but specularly reflected with respect to the temperature axis. In terms of the model, this is achieved if the magnetization of the sublattice A is set, by definition, negative, and that of the sublattice B positive (rather than *vice versa*, as was everywhere assumed above). Then, the temperature dependence of the spontaneous magnetization of each ferrimagnetic grain and the TDM of the ensemble of ferrimagnetic grains will look like those in Fig. 20b (curve *2*), which coincides with the results of experiments [80].

Thus, in terms of the above computerized model of the self-reversal phenomenon, we obtained curves of the TDM which qualitatively correspond to numerous TDM curves with self-reversal, observed in the laboratory and published in the literature. To obtain them, it was sufficient to utilize the single-phase model corresponding to the Néel N type mechanism. Therefore, it is quite possible that the Néel single-phase mechanism of self-reversal of magnetization is widespread in nature. In terms of the model of self-reversal via a two-phase mechanism, which was realized by Krása [69], only one type of the TDM curve was obtained; therefore, this last model is more limited than the model constructed in our works. This indicates that in all registered cases of laboratory-induced self-reversal, even in those where multiphase mechanisms of self-reversal are invoked for the explanation of the observed phenomenon, it is precisely the single-phase N type mechanism that could play the decisive role and cause self-reversal of magnetization.

It should be emphasized that the presence of two mineral phases differing in composition inside ferrimagnetic grains of a rock gives no grounds to state that the self-reversal is caused by the interaction of these phases. The grains can be multiphase, but the self-reversal can be caused by only one phase; therefore, the self-reversal of magnetization can well occur as a result of the action of a single-phase mechanism.

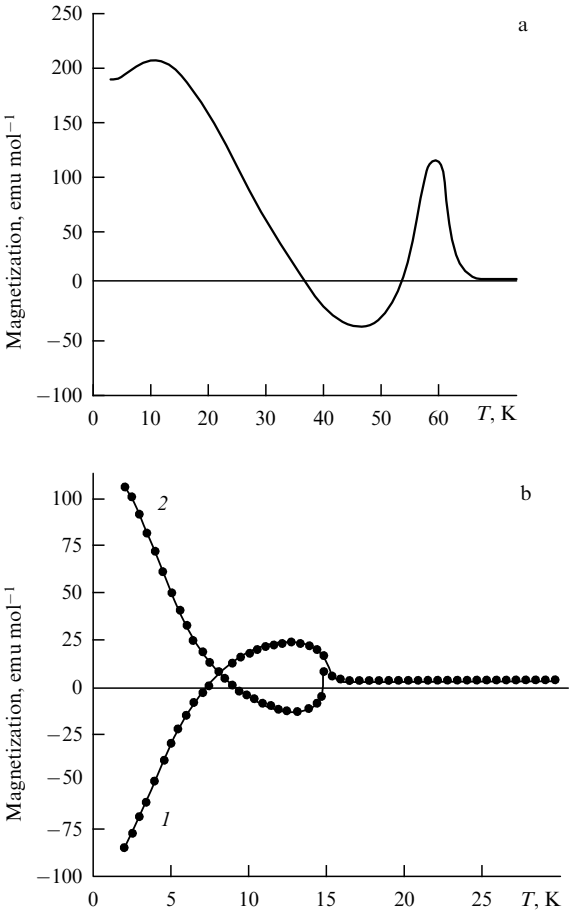

Figure 20. (a) Double reversal of magnetization in modified Berlin blue $(Ni_{0.22}Mn_{0.60}Fe_{0.18})_{1.5}[Cr(CN)_6]\times 7.6H_2O$. (b) Temperature dependences of magnetization of the nickel salt of formic acid $Ni(HCOO)_2\times 2H_2O$: (*1*) on cooling in a magnetic field, and (*2*) on heating of a sample preliminarily cooled in a zero magnetic field. emu stands for electromagnetic unit.

In spite of the acceptable results obtained in terms of the above-constructed computerized model with its description of the self-reversal phenomenon via the Néel N type mechanism, the model can have its restrictions and, certainly, there exist ways of refining the model. For example, it is possible to consider, apart from single-domain grains, an ensemble of pseudo-single-domain grains, or an ensemble of multidomain grains, or even some mixed ensembles, thereby bringing a virtual rock closer to a real one. Other ways of improving the model also most certainly exist.

The debates about the 'true' mechanism of self-reversal of magnetization have been going on for more than a half-century, which is partly related to the very complex composition of rocks: the problem of revealing first a mineral and then magnetic phase responsible for self-reversal of magnetization can by no means always be solved unambiguously. To date, in our opinion, the only sufficiently substantiated mechanism is the Néel N type mechanism suggested above.

5. Self-reversal of magnetization and reversals of the geomagnetic pole. Ecological problems of geomagnetic field reversals

The evolution of the Earth has always occurred in a geomagnetic field, which has exerted effects on both the evolution itself and the properties of the Earth's mass, as well as on the biosphere and humankind. Experiments performed by biophysicists show that animals develop more slowly and are sick more frequently if they are placed in a space shielded from the GMF. All this in full measure refers to humans. It is known that people respond to changes in magnetic activity. Therefore, the magnetic field of the Earth has utmost ecological significance. Moreover, it is the magnetic field that makes possible life itself on Earth and it is only after the appearance of a GMF that there appeared conditions for the generation of life. This phenomenon may be linked to the fact that the Earth's magnetosphere is a natural magnetic trap which captures high-energy particles of solar and cosmic origin and holds them beyond the atmosphere. These trapped particles form radiation belts around the Earth. In the absence of a GMF, part of these particles would be able to reach the Earth's surface and take enormous tolls or even destroy all living matter. However, during GMF reversals, whose duration can reach several thousand years, the strength of the magnetic field is close to zero, i.e., just such a case is realized. Certainly, in the absence of a magnetic field particles of the solar and cosmic radiation can partly be retarded in the upper layers of the atmosphere and partly lose their destroying properties before they reach the Earth's surface. Nevertheless, in these conditions the atmosphere cannot serve as an efficient magnetic trap.

Reversals of the geomagnetic field, during which unpredicted changes in both biotic and abiotic nature on the Earth's surface can occur, represent a kind of revolution which exerts a very strong effect on the course of the evolution of the Earth.

It is for just this reason that studying GMF reversals is a primary problem of modern geophysics. However, as was mentioned in the Introduction, the reversals, even if they are real, cannot be studied directly because of their very long duration. And the only indirect way of studying GMF reversals is the investigation of the processes, mechanisms, and occurrence of the self-reversal of magnetization of rocks. A clear understanding and a detailed development of the self-reversal mechanism in nature will make it possible to estimate the probability of processes of self-reversal, and thereby the probability of GMF reversals. The development of methods of laboratory diagnostics of the mechanism responsible for the formation of a reverse NRM will permit researchers, in each concrete case where it is revealed in nature, to determine whether this reverse NRM is caused by a GMF reversal or self-reversal of magnetization and to refine the commonly accepted scale of reversals by eliminating 'false' reversals, i.e., those that did not exist in reality but were restored from the reverse NRMs of rocks that arose as a result of the processes of self-reversal.

6. Conclusions

We showed in this review that the purely physical problem of self-reversal of magnetization of ferrimagnets can have fundamental global significance for modern geophysics. The problems of the evolution of the geomagnetic field and the evolution of the Earth itself cannot be solved in full measure without refining what information is carried by the reverse NRM of a rock in each concrete case, i.e., whether it is related to GMF reversals or to the internal property of its ferrimagnetic component — the ability of becoming magnetized antiparallel to the GMF. Methods for a sufficiently complete account of cases of self-reversal and

for understanding its physical mechanism should be developed as well.

Determining whether GMF reversals occurred or not is only one of the problems related to geophysical and ecological investigations. As is known [5], the magnetochronological scales of reversals of the geomagnetic field are used for studying global geophysical phenomena, such as the tectonics of lithospheric plates and the formation of mountain massifs and oceans, as well as for studying the structure and properties of the Earth's deep interior, the structure and evolution of the internal and outer layers of the Earth's core, etc.

The determination of the presence or absence of reversals undoubtedly is also of very large significance for the construction of the theory of the origin of the geomagnetic field.

The necessity of improving paleomagnetic methods of investigating the GMF and the interpretation of the results obtained should be noted. In paleomagnetic investigations, the simplest model of the GMF is usually employed, namely, a dipole directed along the axis of the Earth's rotation, with magnetic poles being rigidly tied with the geographic poles. It is assumed that this model is also valid for the ancient GMF of any geological age. Therefore, if the paleomagnetic data based on NRM measurements show that the ancient paleomagnetic pole deviated from the modern geographical pole by an angle $\alpha < \pi$, then, according to the model, the ancient geographic pole also should be deviated at the same angle. However, it is supposed in the paleomagnetic interpretation that no pole deviations occurred but rather a corresponding block of the Earth's crust was shifted. So, there appears a possibility of calculating displacements of continents and lithospheric plates.

If the deviation is just $\alpha \approx \pi$, then it is assumed that the magnetic dipole was turned around by $\alpha \approx \pi$, whereas the geographic pole remained at its place, i.e., the magnetic pole has deviated from the geographic pole by an angle π. This case is interpreted as a GMF reversal. Such an approach to the interpretation of paleomagnetic data is likely to be possible but, in our opinion, it requires a stronger theoretical justification. The more so since, as was shown in this review, there exists a mechanism for the formation of a reverse NRM that is an alternative to the GMF reversals, namely, a spontaneous self-reversal of magnetization I_S of ferrimagnetic minerals of rocks.

At present, the commonly accepted concept in geophysics is that self-reversal of magnetization in rocks does not exert any substantial effect on the number of reversals that have been registered in the World Magnetochronological Scale (magnetic reversal time scale). However, there were already cases where the previously determined reversals proved to be the result of self-reversal of magnetization of rocks [1] [93]. We think that irrespective of how small the number of such cases is, the problem of self-reversal does exist. As was shown in this review, nature is arranged in such a way that ferrimagnetic minerals erupt onto the Earth's surface with magma: these minerals are mainly titanomagnetites, and some of them having a certain composition possess the property of self-reversal. This has been revealed and studied in many laboratories worldwide. Thus, the self-reversal effects have repeatedly been reproduced experimentally in the laboratory. At the same time, we cannot, in principle, directly observe GMF reversals, since their average duration is about 5000 years.

Since we cannot directly study the reversal process, it is expedient to pay greater attention to another process, namely, self-reversal of magnetization, since, by studying the properties and the physical mechanism of this process, we can reach conclusions on the probability of the occurrence of this process in nature and thereby on the occurrence of the alternative process of reversals. A paradoxical statement may be proposed: by studying self-reversal of magnetization in much detail, we make a contribution to the investigation of GMF reversals. As a result, this will permit us to determine the role of reversals in the formation of reversely magnetized rocks.

The determination of the mechanism of each concrete experimentally established case of self-reversal is a very complex problem which has no unambiguous solution. As was shown in this review, many researchers suggest various physical mechanisms for this phenomenon. There exist two basic types of self-reversal mechanisms: (i) due to the interaction (magnetostatic or exchange) of different mineralogical phases in a two-phase or multiphase ferrimagnetic mineral, and (ii) due to a change in the sign of I_S in a single-phase mineral, resulting from changes in the molecular-field constants with temperature or as a result of chemical transformations.

Although the effects of chemical transformations on self-reversal have been considered in several works (see, e.g., Refs [23, 78]), this area has been given insufficient attention. To clearly understand the nature of chemical transformations that can lead to self-reversal of magnetization (except for oxidation, which has been studied well), additional experiments are needed.

The investigations performed in our works [1, 2, 70] and by a number of other authors [72] showed that the most probable physical mechanism of self-reversal of magnetization is the Néel N type mechanism, i.e., the change in the sign of I_S of a two-sublattice ferrimagnet. This is the simplest and most efficient mechanism. The exchange interactions inside individual sublattices and between the sublattices are characterized by large energies, so that it is difficult to imagine some other mechanism of interactions in the mineral, which would be energetically competitive with the Néel N type mechanism.

In our investigations, we presented not only indirect [70] but also direct arguments in favor of the operation of the N type mechanism in the cases of self-reversal studied in our works [1, 97].

In order to expand the possibilities of investigating the curves of the temperature dependence of magnetization of self-reversing samples of rocks, whose number is always limited, we constructed a computerized model of self-reversal of the N type [97].

This model makes it possible, by varying the magnitudes of the spontaneous magnetizations of sublattices and the exchange interactions inside the sublattices and between the

[1] The reverse magnetization of Laschamp and Olby lavas from the Central massif of France (province Auvergne) is assumed to be the first well-documented confirmation of a GMF digression which subsequently was called the Laschamp digression — that is, it is considered as being formed in a GMF of opposite polarity. The authors of Ref. [93] investigated a series of samples from Laschamp and Olby lavas and revealed a self-reversal of the NRM, which permitted them to suppose that the reverse magnetization of these lavas is related to the processes of self-reversal of magnetization rather than to a GMF digression. So, the question arises — has this digression occurred at all?

sublattices, as well as the strength of the applied magnetic fields, to obtain quite varied curves of the temperature-dependent magnetization.

The model permitted us to reproduce virtually all $I_T(T)$ curves available in the literature, including the $I_T(T)$ curves obtained by those researchers who explain the self-reversal cases observed in their works by other physical mechanisms differing from the N type mechanism. This indicates a sufficient universality of our model based on the N type mechanism, and thereby the universality of the N type mechanism itself.

The effect of self-reversal of magnetization in sedimentary rocks requires additional studies, which is connected with certain difficulties, since the sediments are strongly subjected to chemical transformations upon heating. Nevertheless, attempts to study the self-reversal of magnetization in sediments should be continued. This, in particular, will make it possible to compare the self-reversal effects in igneous and sedimentary rocks and to clarify their possible differences. However, in our opinion, the self-reversal of magnetization in sedimentary rocks can also occur via a mechanism of the N type.

To date, the properties of self-reversal of extraterrestrial rocks (in particular, of meteorites) have virtually gone unstudied. Falling meteorites become strongly heated because of friction in the air and after they fall to Earth they are cooled in a GMF, similar to erupted rocks, i.e., they are subject to thermomagnetization. It is possible that the major part of magnetization is acquired by the meteorites in the process of thermomagnetization in a GMF. Therefore, it is logical to suppose that, similar to igneous rocks, meteorites can also reveal properties of self-reversal upon laboratory thermomagnetization or thermodemagnetization. In addition, upon contact with the Earth's surface the meteorites suffer the action of strong impacts and their magnetic characteristics change as compared to those they had prior to falling. At this moment, they can also possibly acquire properties of self-reversal.

To summarize, we can say that the investigation of self-reversal of magnetization has very great scientific importance. The results of such investigations will lead to a more substantial interpretation of paleomagnetic data and a deeper understanding of the evolution of the GMF and the Earth as a whole.

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