ISSN 0027-1349, Moscow University Physics Bulletin, 2010, Vol. 65, No. 3, pp. 209–213. © Allerton Press, Inc., 2010.
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The Puzzles of Natural Ferrimagnetics

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Received February 25, 2010

Abstract—Paleomagnetic studies have revealed that natural ferrimagnetics, which are constituents of igneous rocks, can be magnetized antiparallel to the magnetizing field. This phenomenon is called self-reversal of magnetization. The question of the actual existence of inversions of the geomagnetic field is still under investigation. This paper considers the physics of the anomalous behavior of natural ferrimagnetic thermomagnetization (TM). The existence of thermomagnetization arising in natural ferrites against the direction of a magnetizing field is experimentally confirmed. It is shown that even after the demagnetization of a ferrimagnetic by alternating magnetic field a significant increase in residual magnetization can occur when it is heated in the absence of a magnetic field.

DOI: 10.3103/S0027134910030100

INTRODUCTION

The geomagnetic field (GMF) has played a huge role in the evolution of our planet [1, 2].

The GMF permeates the entire Earth, ocean, and atmosphere, affects animate and inanimate nature, and also magnetizes rocks. Interacting with the solar wind in terrestrial space, i.e., with the plasma flow that moves in a radial direction from the Sun, the GMF forms the magnetosphere [3]. The magnetosphere and the radiation belts that are situated in the upper layers of the atmosphere protect the Earth's surface from the strong radiation of the solar wind and cosmic rays. This promoted the development of the primary elements necessary for life. The presence of GMF lines near the Earth's surface established the anisotropy that was crucial for the beginnings of living elements. The GMF may have influenced not only the beginnings, but also the development of life on Earth, because life developed under the protection of the magnetosphere.

Paleomagnetic investigations, which began in the middle of the 20th century, have shown that natural remanent magnetization (NRM) of ancient rocks may be directed along GMF directions (straight NRM), as well as in the opposite direction (inverse NRM) [3].

It has been suggested [3] that inverse NRM that is approximately antiparallel to the present-day GMF has been created in the GMF. Consequently, the GMF changes its polarity periodically and so-called inversions of the GMF have occurred. According to the paleomagnetic data, more than 1000 inversions have occurred in the last 400–600 million years; the duration of the change in the GMF direction is about 5000 years. During the GMF polarity inversion its intensity could be nearly zero, and the magnetosphere could be partly or completely destroyed. It is possible that in the absence of the magnetosphere the Earth's surface was exposed to strong radiation, which had an adverse effect on the biosphere and life on the Earth.

The existence of GMF inversions has as yet only indirect evidence—the inverse NRM of rocks. Moreover, in laboratory studies of ferrites and rocks numerous examples of magnetization antiparallel to the magnetizing field have been found [4]. This phenomenon is called self-reversal of magnetization. Solving the problem of reverse NRM is of global significance for understanding the evolution of the Earth and GMF, because the inverse NRM of rocks may be related either to GMF inversions or to the self-reversal of magnetization phenomenon.

FORMULATION OF THE PROBLEM

1. Theory of Ferrimagnetism by L. Neel

Ferrimagnetics are magnetically ordered substances with two or more magnetic sublattices. The most abundant ferrimagnetics, magnetite and titanomagnetite, as well as hemoilmenite, usually have two magnetic sublattices.

In a two-sublattice (*A* and *B*) ferrite there are two spontaneous magnetizations I_{AS} and I_{BS}, directed oppositely to each other. Summary spontaneous magnetization is $I_S = I_{BS} - I_{AS}$ or $I_S = I_{AS} - I_{BS}$ [5].

The basic types of dependences $I_S(T)$ for ferrimagnetics calculated by *L*. Neel are shown in Fig. 1 [6]. These dependences have a very complicated character. For example, with increasing T (dependences of types *M* and *P* in Figs. 1 and 2a) magnetization $I_S(T)$ does not decrease, as it would in the case of ferromagnetics, but increases. In the dependences $I_S(T)$ there can be points of compensation T_K, when at $T = T_K$ magnetizations of the sublattices are equal $I_{AS} = I_{BS}$, and $I_s(T_k) = 0$. Moreover, at temperatures above or below

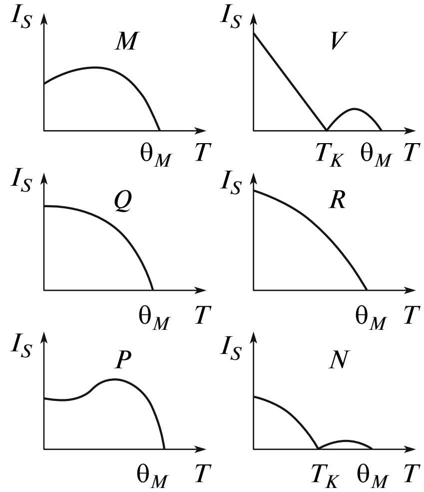

Fig. 1. Basic temperature dependence types for spontaneous magnetization of ferrites [6].

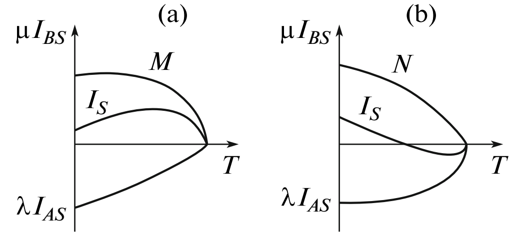

Fig. 2. Possible dependences $I_S(T)$ at different temperature behaviors of I_{AS} and I_{BS} [4].

T_K the magnetization I_S can be directed antiparallel to the magnetizing field (dependences of types V and N in Fig. 2b).

As a matter of fact, L. Neel divided ferrites into two groups, depending on the character of the dependence $I_S(T)$ [6]. In the first group $I_S(T)$ is the same function of T (types Q and R in Fig. 1), as the dependence $I_s(T)$ for ferromagnetics. Ferrites, which possess dependences $I_S(T)$ of types N and V, belong to the second group. Self-reversal of magnetization can be observed for ferrites from the second group. In the monograph by S.V. Vonsovskii [16] other, quite complex anomalous effects of dependences $I_S(T)$ for ferrimagnetics, are also discussed.

In our opinion, the main features of self-reversal of magnetization can best be described in terms of a change in sign of $I_S = I_{BS} - I_{AS}$, which occur due to the different temperature dependences of sublattices A and B magnetizations (dependences $I_S(T)$ of types V and N).

The authors of [4, 7] established that the most likely cause of self-reversal of magnetization in nature is the N type dependence of $I_S(T)$ (Fig. 1 and 2b).

Consider the conditions in a two-sublattice ferrimagnetic that lead to a change in the sign of I_S with changing T [13]. The spontaneous magnetizations I_{AS} and I_{BS} of sublattices A and B can be expressed [13] as:

$$I_{AS} = \lambda I_\infty B_J\left(\frac{Jg\mu_B n(\alpha I_{AS} - I_{BS})}{kT}\right),$$
$$I_{BS} = \mu I_\infty B_J\left(\frac{Jg\mu_B n(\beta I_{BS} - I_{AS})}{kT}\right). \quad (1)$$

The summary magnetization I_S is the difference between the sublattice magnetizations:

$$I_S(T) = I_{BS}(T) - I_{AS}(T). \quad (2)$$

In (1), λ and μ are the fractions of magnetic ions in the sublattices A and B ($\lambda + \mu = 1$); $B_J(\alpha)$ is the Brillouin function; J is the internal quantum number; g is the Lande g-factor; μ_B is the Bohr magneton; and k is the Boltzmann constant.

In the general case, values of I_∞, J and g may be different for different sublattices. However, we can ignore this fact in our case.

The expressions in the arguments of the functions $B_J(\alpha)$:

$$n(\alpha I_{AS} - I_{BS}) = H_{MA}$$
$$n(\beta I_{BS} - I_{AS}) = H_{MB} \quad (3)$$

are molecular fields that exist inside the sublattices A and B, respectively. The values n, $n\lambda$, and $n\beta$ are constants of the molecular field, which determine the interactions of magnetic moments between the sublattices and inside them.

The anomalous dependence $I_S(T)$ is characterized by a change in the sign of I_S, while T changes and reaches $T = T_K$. Assume that $\mu > \lambda$. As the argument of the Brillouin function $\alpha \gg 1$ and $B_J(\alpha) \approx 1$ at $T \longrightarrow 0$ K, the next correlation is true, according to (1):

$$|I_{BS}| > |I_{AS}| \text{ and } I_S = I_{BS} - I_{AS} > 0. \quad (4)$$

When the sign of I_S changes with increasing T at temperatures close to the Curie points T_c the next correlations must be fulfilled (with λ and μ being constant):

$$|I_{AS}| > |I_{BS}| \text{ and } I_S = I_{BS} - I_{AS} < 0. \quad (5)$$

Let us examine when this is possible. At $T \longrightarrow T_c$ (T_c is the Curie point), the argument $\alpha \ll 1$ and the Brillouin function is written [13] as:

$$B_J(\alpha) = \frac{J+1}{3J}\alpha. \quad (6)$$

Using (6),we can obtain the following expressions for I_{AS} and I_{BS} at $\alpha \ll 1$ from (1):

$$I_{AS} \approx \frac{Cn}{T}\lambda(\alpha I_{AS} - I_{BS}),$$
$$I_{BS} \approx \frac{Cn}{T}\mu(\beta I_{BS} - I_{AS}), \quad (7)$$

where $C = \frac{NJ(J+1)g^2\mu_B^2}{3k}$, and N is the Avogadro constant.

At $T = T_K$, where the sign of I_s changes, the equality $I_{BS} - I_{AS} = 0$ is true, which is written using (7) as:

$$I_S = \frac{Cn}{T}[\mu(\beta I_{BS} - I_{AS}) - \lambda(\alpha I_{AS} - I_{BS})] = 0.$$

Considering that at $T = T_K$ the values $|I_{AS}|$ and $|I_{BS}|$ are equal, we obtain the expression for the boundary between ferrimagnetic phases with opposite directions of I_S:

$$\lambda(\alpha + 1) - \mu(\beta + 1) = 0. \quad (8)$$

The left side of (8) will be positive when $I_S > 0$, and negative when $I_S < 0$. For the latter case we arrive at the following condition:

$$\frac{\beta + 1}{\alpha + 1} > \frac{\lambda}{\mu}. \quad (9)$$

This is a fulfillment of (9), which determines the change in sign of I_S for a temperature increase from 0 K to T_c, which leads to N-type curves $I_S(T)$ at $T = T_K$.

The considered theory shows that a compensation point must appear on the N-type curves. However, T_K will be observed in experiments only for a homogeneous ferrimagnetic having a single Curie point in its phase transition. In fact, a ferrimagnetic grain ensemble in a rock possesses a diffuse phase transition $T_c \pm \Delta T_c$, which results in a diffuse T_K value; therefore, in strong enough fields H the curve $I_S(T)$ will not reach zero and one can observe only some decrease in I_S near $T_K \pm \Delta T_K$ [17].

2. Indirect Exchange

As is well known, the exchange interactions (molecular fields) E_A and E_B act in the sublattices of ferrites, which orders the magnetic moments in the sublattices. There is also an exchange interaction E_{AB} between the sublattices. The energy E_{AB} between spins of neighboring metal ions is negative and thus spins are arranged in antiparallel order.

In ferrimagnetics metal ions (cations) are detached from each other unequally by negative ions (O^{2-}). The distance between metal ions is usually too large for direct exchanges to occur, which usually act at distances ~10^{-8} cm. Therefore, in ferrite-like substances a significant role in exchange interactions belongs to intermediate anions (superexchange or indirect exchange).

Due to this, very complex interactions can arise between ions in the sublattices of ferrimagnetics. A suggestion was made by K.P. Belov [10] that the magnitudes of exchange interactions in ferrite sublattices can differ substantially. In connection with this, Belov introduces the concept of a "weakly ordered" or "weak" magnetic sublattice.

According to [10], the anomalous dependences $I_S(T)$ of Neel types N, M, P can be explained with the help of the "weak" sublattice concept, including the presence of a compensation point T_K, when $I_{AS} = I_{BS}$, and other anomalous dependences $I_S(T)$ in ferrites. Successful interpretation of anomalous effects using the concept of a "weak" magnetic sublattice in studies of ferrites was performed in [11, 12].

3. Diffusion Anisotropy

We studied continental and underwater oceanic rocks that contain natural ferrites (magnetite and titanomagnetite) in the form of small grains that are chaotically embedded in the basalt matrix. But in studies of magnetic properties of rocks that contain natural ferrites we consider the sample as a single massive ferrite rather than separate particles, scattered in a basalt sample.

Such a possibility is explained by the fact that ferrites and ferrite particles undergo annealing in the geomagnetic field during their natural magnetization. It is known that diffusion uniaxial magnetic anisotropy in ferrimagnet particles can arise during such annealing [5]. The same anisotropy also appears in the laboratory thermomagnetization of rock samples. The energy of uniaxial anisotropy can significantly exceed the energy of crystallographic anisotropy and the magnetic field direction becomes the single easy magnetization direction in all ferrimagnetic particles. This allows us to consider a rock sample with ferrite particles as a single ferrite sample.

The theory of diffusion uniaxial anisotropy was created by Chikazumi [13]. It was developed in works by Neel [14] and Toniguchi and Yamamoto [15].

EXPERIMENTAL RESULTS

The authors carried out a number of experiments on underwater basalt samples in order to investigate the anomalous properties of natural ferrimagnetics.

New anomalies in the behavior of ferrimagnetics that are cemented in basalts were experimentally found under laboratory thermomagnetization and demagnetization of underwater oceanic basalt samples in [11].

We performed successive partial thermomagnetizations (PTMs), which were generated in different temperature intervals on a sample of underwater basalt. The sample was then cooled in the absence of a magnetic field to the liquid nitrogen temperature. Then the change in magnetization upon the heating of the sample to $T \approx 300°C$, also in the absence of a magnetic field, was recorded (Fig. 3). It is evident that the intensities of all PTMs did not decrease, but rather increased, as T grows. Their increase continued up to temperatures near 300°C.

The increase in magnetization on heating in the absence of a magnetic field can be accounted for by

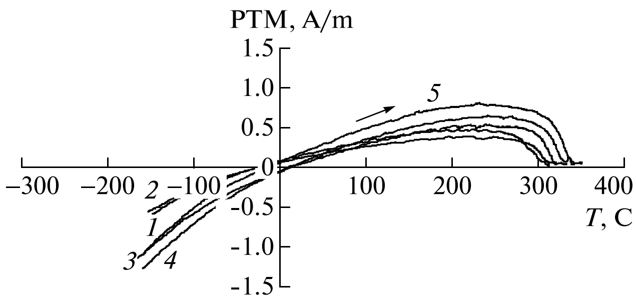

Fig. 3. Changes in PTM ($H = 0.1$ mT), created in different temperature ranges, when heating the sample 29/1(2) in the absence of a field: (*1*) $T = 300–275°C$; (*2*) $T = 310–300°C$; and (*3*) $T = 320–310°C$.

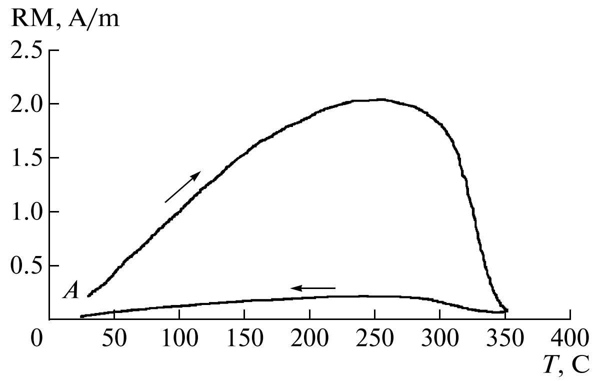

Fig. 4. Change in magnetization of the sample 29/1(2), starting from zero state *A*, on heating and cooling in the absence of a field. The zero state *A* was obtained using demagnetization of PTM (350–300°C, 0.1 mT) by alternating magnetic field of $h \approx 0.1$ T.

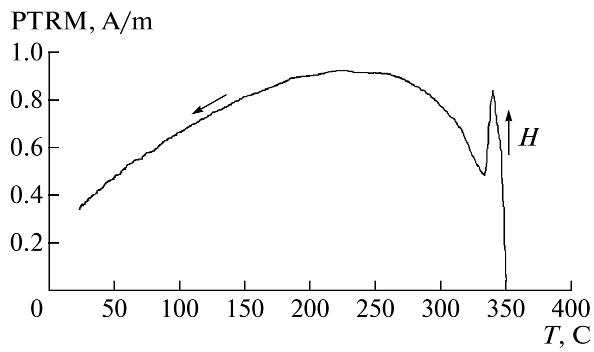

Fig. 5. Arising PTM in sample 29/1(2) in the temperature range 350–340°C in a field of $H = 0.1$ mT; in the range $T = 340–20°C$, $H = 0$ [11].

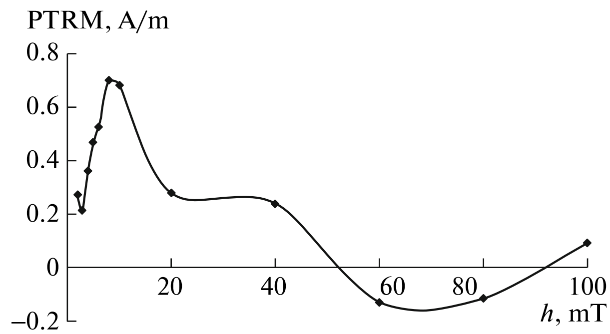

Fig. 6. Demagnetization of PTM ($T = 325–300$, $H = 0.1$ mT) of sample 16/30 by an alternating field.

the suggestion that one of the ferrimagnetic sublattices in the basalt sample is "weak" and has substantially weaker exchange energy, which orders the magnetic moments in the sublattice. On heating this ordering is destroyed, which leads to an increase in the summary spontaneous magnetization of the sample due to the I_s of the strong sublattice. At temperatures >270°C the magnetic ordering of the second sublattice collapses as well, and this results in a decrease in the summary magnetization of the sample to zero.

The same effect of an increase in magnetization upon the growth *T* and $H = 0$ was obtained with the same sample when PTM (350–300°C) was demagnetized before heating by means of alternating magnetic filed of $h \approx 1000$ Oe (0.1 mT) (Fig. 4).

Other anomalous effects in the behavior of natural ferrites are also presented in [11]. The complex character of a forming PTM is shown in Fig. 5. The abrupt jump of magnetization at $T = 350°C$ is notable in this figure. This is apparently due to the full ordering of one of the ferrimagnetic sublattices at $T = 350°C$, which involves the formation of large magnetization along the *H* direction. The abrupt decrease in magnetization is due to the shutdown of the magnetic field. The further magnetization change from $T = 325°C$ and $T = 20°C$ complies with the Neel's type *M* dependence of $I_S(T)$ (Fig. 1).

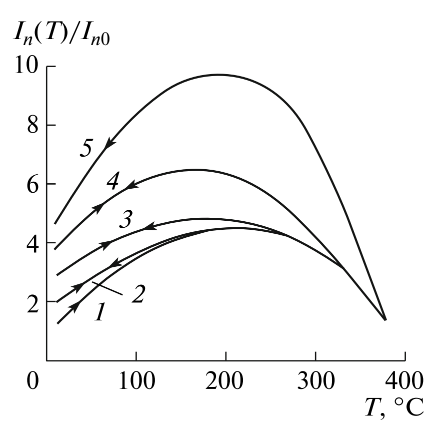

Fig. 7. Cyclic demagnetization I_n of a sample of transitional basalt [9].

The complex changes in the behaviors of spontaneous magnetizations $I_{AS}(T)$ and $I_{BS}(T)$ leads to a complex construction of thermomagnetization, arising at changing T. One indication of this is the very unusual PTM demagnetization curve that occurs in an alternating magnetic field h (Fig. 6).

Anomalous temperature dependences of I_n were obtained in [9] in a study of a transitional basalt sample (Fig. 7).

CONCLUSIONS

The data on different anomalous processes of ferrites magnetization that are given in the present paper allow us to suggest that the thermomagnetization direction in rock samples that have ferrite properties does not necessarily reflect the magnetizing field direction. Additional detailed physical research on the thermomagnetization of a specific ferrite sample is needed, at the least, in order to determine whether the TRM or NRM direction corresponds to the magnetic or geomagnetic field direction for which these magnetizations were created.

In view of the observed anomalous cases of ferrite magnetization, the long-standing question "inversion or self-reversal of magnetization?" becomes much more complicated. The above data on the anomalous magnetization of ferrites allow us to conclude that the problem of inverse NRM can be solved only by detailed investigation of ferrimagnetic minerals that are constituents of ancient rocks; it is necessary to prove in every case that the magnetic properties of ferrimagnetics that are parts of a rock are analogous to the properties of ferromagnetics, i.e., that they have temperature dependences $I_S(T)$ of Neel's type Q or R.

ACKNOWLEDGEMENTS

The work was supported by the Russian Foundation for Basic Research, project no. 08-05-00623.

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