Low Temperature Characteristics of Rock Magnetism

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Abstract

Since the magnetocrystalline anisotropy constants \( K_1, K_2 \) and the magnetostriction coefficients \( \lambda_{100}, \lambda_{111} \) of titanomagnetites change appreciably with temperature in the low temperature range, the magnetic susceptibility and remanent magnetization of these minerals depending on these parameters also change markedly with temperature. In particular, the magnetite-rich titanomagnetite has the isotropic point, at which \( K_1 = 0 \), at a low temperature. The marked reduction of magnetocrystalline anisotropy energy around the isotropic point results in some remarkable magnetic phenomena. These phenomena are:

(a) On cooling, IRM almost vanishes or is sharply reduced at the isotropic temperature, and on subsequent heating, once diminished magnetization is recovered to a certain extent above the isotropic temperature;

(b) By heating from a temperature below the isotropic point to the room temperature in a magnetic field \( (H) \), the sample acquires remanent magnetization;

(c) As IRM almost vanishes at the isotropic temperature on cooling and its recovery on heating is of a small amount \( (< \frac{1}{5}) \) while TRM's recovery rate is 80%–90%, the procedure of cooling down a rock sample to a temperature below the isotropic point can work as an effective method of cleaning IRM and VRM of magnetites and titanomagnetites.

1. Introduction

Remanent magnetization of rocks are subject, in general, to three major factors of magnetic minerals, i.e. the magnetocrystalline anisotropy, the stress-induced anisotropy and the shape anisotropy. For the sake of simplicity, we may consider a case of uniaxial anisotropy of a single domain particle for a rough estimation of the effect of these three factors. Then, the anisotropy constant and the spontaneous magnetization being denoted by \( K \) and \( J_s \), respectively, the coercive force \( H_c \) is expressed as

\[
H_c = \frac{2K}{J_s}.
\]  

In case of the magnetocrystalline anisotropy alone, we may take the magnetocrystalline anisotropy constant of magnetite as a representative value for \( K = K_1 = -1.4 \times 10^5 \text{erg/cm}^3 \) at the room temperature.

The anisotropy constant \( (K_s) \) caused by internal mechanical stress \( \sigma \) is given as

\[
K = K_s = \frac{3}{2} \lambda \sigma,
\]

where \( \lambda \) means the mean magnetostriction coefficient. For magnetite, \( \lambda_{100} = -20 \times 10^{-6} \) c.g.s. and \( \lambda_{111} = 78 \times 10^{-6} \) c.g.s. at the room temperature. The magnitude of \( \sigma \) in magnetic mineral
grains is estimated to be $10^9$ c.g.s in the order of magnitude. Taking the mean value of $\lambda_{100}$ and $\lambda_{111}$ as $\lambda$, (2) gives that $K = 8 \times 10^4$ erg/cm$^3$.

On the other hand, the shape anisotropy constant ($K_2$) for an ellipsoid of $M$ and $N$ respectively in the demagnetizing factors along the minor and major axes is expressed as

$$K = K_2 = \frac{1}{2} (M-N)/J_s^2. \quad (3)$$

Since $J_s \approx 500$ c.g.s.e.m.u. and $M-N$ may be of the order of unity, $K$ in this case is $10^5$ erg/cm$^3$ in the order of magnitude.

We may consider therefore that the contributions of the these factors to the magnetic anisotropy are roughly the same at the room temperature.

As well known, the magnetocrystalline anisotropy constant $K_1$ of magnetite becomes zero at about $-140^\circ$C, which is called the isotropic temperature. Above the isotropic temperature, $K_1$ takes negative value, while below the isotropic temperature $K_1$ takes positive value. According to Syôno (1964), $K_1$ of titanomagnetite expressed as $x\text{Fe}_2\text{TiO}_4, (1-x)\text{Fe}_3\text{O}_4$, becomes zero at a certain low temperature for $x = \frac{1}{2}$. The isotropic temperature of titanomagnetite for $\frac{1}{2} > x > 0$ is lower than that of pure magnetite. For the range of $x > \frac{1}{2}$, however, $K_1$ increases exponentially with decreasing temperature, becoming about $10^7$ erg/cm$^3$ at $80^\circ$K.

For the range of $x < \frac{1}{2}$, the magnetostriction coefficient $\lambda_{111}$ decreases with decreasing temperature, and $\lambda_{100}$ is approximately independent of temperature for $x < \frac{1}{6}$ but it increases with decreasing temperature for $\frac{1}{6} < x < \frac{1}{2}$. For the range of $x > \frac{1}{2}, \lambda_{100}$ increases markedly with decreasing temperature, becoming about $10^{-8}$ c.g.s. at $80^\circ$K.

Table I is a reproduction of the main results of Syôno's measurement of $(K_1, K_2)$ and $(\lambda_{100}, \lambda_{111})$ of single crystals of titanomagnetics.

For the low temperature range between the liquid nitrogen temperature ($-196^\circ$C) and the room temperature, and for the magnetite-rich titanomagnetics ($x < \frac{1}{2}$), $K_1$ only varies

<table>
<thead>
<tr>
<th>$x$</th>
<th>$\theta^\circ$C</th>
<th>$K_1$ 10$^5$ erg/cm$^3$</th>
<th>$K_2$ 10$^5$ erg/cm$^3$</th>
<th>$M_s$ emu g</th>
<th>$K_1/M_s$ at 290$^\circ$K</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00</td>
<td>575</td>
<td>$-1.36$</td>
<td>$-1.50$</td>
<td>$-0.40$</td>
<td>$-0.44$</td>
</tr>
<tr>
<td>0.04</td>
<td>550</td>
<td>$-1.94$</td>
<td>$-2.24$</td>
<td>$-1.10$</td>
<td>$-0.18$</td>
</tr>
<tr>
<td>0.10</td>
<td>520</td>
<td>$-2.50$</td>
<td>$-2.90$</td>
<td>$-1.98$</td>
<td>0.23</td>
</tr>
<tr>
<td>0.18</td>
<td>470</td>
<td>$-1.92$</td>
<td>$-2.44$</td>
<td>$-2.04$</td>
<td>$-0.66$</td>
</tr>
<tr>
<td>0.31</td>
<td>400</td>
<td>$-1.81$</td>
<td>$-2.45$</td>
<td>$-1.60$</td>
<td>$-0.10$</td>
</tr>
<tr>
<td>0.56</td>
<td>170</td>
<td>$0.70$</td>
<td>1.19</td>
<td>(25)</td>
<td>(74)</td>
</tr>
<tr>
<td>0.68</td>
<td>80</td>
<td>0.18</td>
<td>4.9</td>
<td>(40)</td>
<td>(120)</td>
</tr>
</tbody>
</table>
in an absolutely marked way, while \( 2 \lambda = \lambda_{100} + \lambda_{111} \) or the directly observed values of magnetostriction coefficient of polycrystalline titanomagnetite are always positive.

In other words, \( K_1 \) only varies markedly from positive values on lower temperature side to negative values on higher temperature side, crossing the isotropic temperature, while both \( K_a \) given by (2) and \( K_r \) given by (3) are kept positive throughout the temperature range.

If, for example, \( H \) depends entirely on \( K_1 \) only, then \( H \) should become zero at the isotropic temperature. It would mean that the remanent magnetization maintained by the coercivity against the demagnetizing field must diminish at the isotropic temperature provided no external magnetic field is applied. On the other hand, the magnetic susceptibility \( \chi \), which can be expressed as

\[
\chi = J_s^2 / 2K \tag{4}
\]

in case of the simplified uniaxial model of a single domain particle, should become extremely large at the isotropic temperature. Hence, if a magnetic field is applied on a magnetite-rich titanomagnetite sample at its isotropic temperature and then temperature is raised up in the field, the increase in \( H_e \) with increasing temperature may result in blocking of a certain portion of magnetization acquired around the isotropic temperature.

If, on the contrary, either the stress-induced anisotropy or the shape anisotropy is dominant, such a remarkable irreversible phenomenon may not take place. This paper is a review of recent experimental works on low temperature characteristics of magnetization of magnetite-rich titanomagnetite and rocks, based on the general idea discussed above, carried out in the writer’s laboratories at Tokyo and at Pittsburgh.

2. Low temperature behaviours of remanent magnetization of titanomagnetite.

When a specimen of magnetite-rich titanomagnetite is cooled down from the room temperature in non-magnetic space, its IRM acquired at the room temperature decreases sharply around the isotropic temperature. On heating of this specimen in non-magnetic space, a certain portion of remanent magnetization is recovered, as illustrated is Fig. 1-(a) for example. In some cases, the intensity of remanent magnetization steps down around the isotropic temperature on the heating process in non-magnetic space, as shown by Fig 1-(b).

* The values in bracket are only lower limit ones on account of under saturation.
** \( \lambda_a \) is the saturation magnetostriction for polycrystalline specimens.

(after Y. Syōno)
Two typical examples shown in Fig. 1 may suggest that the remanent magnetization consists of two components, A and B. A is very sensitive to and dependent mostly on the magnetocrystalline anisotropy $K_1$, while B is almost independent of $K_1$ but is dependent on either $\sigma$ or the shape anisotropy. It will be an essential condition for the A-component that the direction of easy magnetization is along the [111] equivalent axes at temperatures above the isotropic temperature ($T_a$) and is along the [100] equivalent axes below $T_a$.

On cooling in non-magnetic space, the A component may almost vanish at $T_a$, while the B-component remains even below $T_a$. We must, however, take into consideration the magnetostatic interaction between A and B components and among various domains having different values of microscopic coercivity ($h_c$) (Ozima and Nagata 1963) even in the group of A component. Since the B component magnetization acquired at the room temperature is, in general, along the [111] equivalent directions, the magnetostatic interaction between the A and B components in multidomain grains is considered to be much more effective for the A-component magnetization of [111] alignment above $T_c$ than for that of [100] alignment below $T_c$. There are two kinds of magnetostatic interaction, i.e. positive and negative interactions. (Uyeda 1958, Nagata 1961) according to geometrical configurations of distributions of A and B domains. In case of the positive interaction, the effect of B-component magnetization results in the A-component magnetization parallel to the B-magnetization, while in case of the negative interaction it results in the anti-parallel magnetization of A-component.

The above would be the simplest interpretation of the two types of recovery phenomenon shown in Fig. 1. A qualitative justification of the above-mentioned interpretation will be given by the following experimental facts. When a magnetite-rich titanomagnetite sample of polycrystals (and consequently multi-domains) is magnetized at a temperature below $T_c$, the IRM thus acquired is reduced markedly by crossing over $T_c$ on heating in non-magnetic space, but a part of the reduced IRM is recovered on cooling down below $T_c$ in non-magnetic space (Nagata, Kobayasi and Fuller, 1964). The IRM produced at a temperature above $T_c$ and that produced at a temperature below $T_c$ may be called respectively the
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ordinary IRM and the low temperature IRM. Then, the ordinary IRM and the low temperature IRM are inversely related to each other. Namely, when an intense low temperature IRM is put on a sample which had the ordinary IRM above $T_e$, the ordinary IRM is not recovered on heating, the low temperature IRM only being recovered on re-cooling; on the other hand, when an intense ordinary IRM is put on a sample which had the low temperature IRM below $T_e$, the ordinary temperature IRM only is recovered on re-heating. If the secondly-put IRM is not intense enough, both IRMs show their recoveries. In this case, $\gamma_e$ and $\gamma_o$ denoting respectively the recovery ratios of the low-temperature and ordinary IRMs, there holds an approximate relationship that

$$a\gamma_e + b\gamma_o \approx C,$$

where $C$ is a constant and $a = C/\text{maximum value of } \gamma_e$ (corresponding to $\gamma_o = 0$), and $b = C/\text{maximum value of } \gamma_o$ (corresponding to $\gamma_e = 0$). (5) may indicate that there is an inverse relationship between the recoveries of the ordinary and low-temperature IRMs. This experimental fact seems to suggest that there are a number of nuclei of spontaneous magnetization (probably of single domain introduced by the local accumulation of internal stress), whose magnetic fields are favourable to magnetization of environing domains either along the [111] axes or the [100] axes. When all nuclei are magnetized along the [111] axes, only the ordinary IRM can be recovered, no low-temperature IRM being recovered, while when all nuclei are magnetized along the [100] axes, the low temperature IRM only can be recovered.

Mitsuko Ozima (1965), however, has suggested that the magnetostatic coupling may take place also between various domains having different values of microscopic coercivity ($h_c$) of A-component in case of polycrystalline titanomagnetite just as in case of their single crystals, which will be discussed in Section 5.

In any case, the rapid reduction of the ordinary IRM on cooling and that of the low-temperature IRM on heating are considered to take place around the isotopic temperature $T_e$. Full circles in Fig 2 show the temperature of maximum rate of change in IRM with temperature for magnetite-rich titanomagnetites having different contents of ulvöspinel. Hollow circles in the figure are the isotropic temperature of magnetite-rich titanomagnetites obtained from the $K_1 = 0$ condition by Syôno (1964). The agreement between the two sets of independent estimations will be considered satisfactory.

3. Recovery rates of IRM and TRM after cooling.

Let the intensity of the ordinary IRM and that of its residual magnetization after cooling to a temperature below $T_e$ and after re-heating to the room temperature in non-magnetic space be denoted by $J_o (\text{IRM})$ and $J_r (\text{IRM})$ respectively.
Then, the ratio of $J_r(IRM)$ to $J_0(IRM)$ may be defined as the recovery rate of the ordinary IRM for the cooling procedure, being denoted by $r(IRM)$. The amount of $r(IRM)$ increases with increase in the initial value of $J_0(IRM)$, or in other words, with increase in the intensity ($H$) of magnetic field applied for acquiring the IRM, as illustrated in Fig. 3.

Let, on the other hand, the intensity of TRM of the same sample at the room temperature and that of its residual magnetization after cooling to a temperature below $T_c$ and after re-warming to the room temperature in non-magnetic space be denoted by $J_0(TRM)$ and $J_f(TRM)$. Then, the recovery ratio of the TRM will be defined as $r(TRM) = J_f(TRM)/J_0(TRM)$. As shown in Fig. 3, $r(TRM)$ is nearly unity for small values of $J_0(TRM)$ and it decreases gradually with increase in $J_0(TRM)$, or with increase in $H$. It seems likely that $r(IRM)$ and $r(TRM)$ approach to a same asymptotic value for sufficiently large values of $H$.

The experimental result represented by Fig. 3 may indicate that the coercivity to maintain IRM acquired in a weak magnetic field is largely due to the magnetocrystalline anisotropy, while the coercivity to maintain TRM produced in a weak field is mostly due to the stress-induced anisotropy or the shape anisotropy, and furthere that both the magnetocrystalline anisotropy and the non-magnetocrystalline anisotropy become involved in the coercivity of both IRM and TRM acquired in a strong magnetic field.

The definite difference in the recovery ratios of IRM and TRM for the low temperature treatment may be used for the creaning of remanent magnetization of igneous rocks. Because natural remanent magnetization of igneous rocks is believed to be due mostly to TRM and IRM produced in a weak geomagnetic field. When a specimen of igneuous rock is cooled, for example, in liquid nitrogen, its remanence owing to IRM mechanism should almost vanish while its remanence owing to TRM mechanism should remain almost inviably. Mitsuko Ozima (1964) has shown that this low-temperature cleaning method is practically effective for granitic rocks.

4. Inversed-type Thermoremanent Magnetization.

The magnetic susceptibility of magnetite-rich titanomagnetite, in which the magnetocrystalline anisotropy is dominant, shows an anomalous increase around the isotropic temperature ($T_c$) because of small values of $K_i$.

When such a sample is heated in a magnetic field $H$ from a temperature below $T_c$ to the room temperature, the magnetization acquired around $T_c$ will be blocked by increasing coercivity caused by increasing temperature. Nagata, Ozima and Yama-ai (1963) experimen-
tally demonstrated such an acquisition of stable remanent magnetization. Fig. 4 illustrates a sharp decrease of coercive force \( (H_c) \) of magnetite around its isotropic temperature. Fig. 5 shows partial thermo-remanent magnetizations of inversed type acquired by applying \( H \) during temperature ranges of 20 degrees in interval on heating of magnetite from the liquid nitrogen temperature. As shown in Fig. 5 the intensity of partial TRM of inversed type at the room temperature takes the maximum value around \( T_c \). Obviously, the partial TRM of inversed type thus defined includes the IRM obtained in the temperature range where \( H \) is applied. A comparison of Fig. 5 with Fig. 4 may indicate an approximate relation that (intensity of PTRM of inversed type) \( \times H_c \equiv \) constant.

The plausibility of this relationship has been theoretically proved by Ozima and Nagata.
The basic idea of their theoretical interpretation is as follows: Introducing the concept of microscopic coercive force \( h_c \) of individual magnetic domains having \( j \) of spontaneous magnetization, the intensity \( J_r \) of IRM produced in a magnetic field \( H \) is given as
\[
J_r = \int_0^H \frac{j \cdot n(h_c) dh_c}{N j_f}, \quad (6)
\]
where \( N \) and \( n(h_c) \) denote respectively the bulk demagnetizing factor and the distribution function of microscopic coercivity in the concerned assemblage of individual domains. The bulk coercive force \( H_c \) can then be expressed as
\[
H_c = \frac{1}{2} \left( h_c \right)_m \quad (7)
\]
For a rough approximation, \( n(h_c) \) will be given as \( n(h_c) = n_o \) for \( 0 \leq h_c \leq (h_c)_m \) and \( n(h_c) = 0 \) for \( (h_c)_m < h_c \). Then (7) becomes
\[
H_c = \frac{1}{2} \left( h_c \right)_m \quad (8)
\]
and (6) is expressed as
\[
J_r = \frac{j n_o H}{1 + N n_o}. \quad (9)
\]
Since \( n_o (h_c)_m = \text{constant} = N_o \), and consequently \( n_o = N_o / 2 H_c \), (9) is expressed as
\[
J_r = \frac{j N_o}{1 + N N_o / 2 H_c} \cdot \frac{H}{2H_c}, \quad (10)
\]
Thus, \( J_r \) is, roughly speaking, inversely proportional to \( H_c \). The larger remanence thus acquired at a low-temperature for which \( H_c \) is smaller becomes blocked at a higher temperature for which \( H_c \) is larger.

5. Reversal of IRM of single crystal of magnetite at low temperatures.

Fig. 6 shows an example of change with temperature of the intensity of IRM of a single crystal of magnetite acquired at the room temperature. The IRM sharply decreases around the isotropic temperature on cooling in non-magnetic space and is partly recovered on reheating process. The particular fact in this case is that the magnetization below \( T_c \) is opposite to that above \( T_c \). The reversal of IRM of a magnetite single crystal at low temperatures below \( T_c \) can take place for both [100] and [111] directions. (Yama-ai, Ozima and Nagata 1963) As shown in Fig. 7, however, the condition for the reversal is particularly sensitive to the dimension ratio of measured samples; namely, the reversal can take place only in samples of small dimension ratio, in which the bulk demagnetizing factor is large.

It will be presumed, therefore, that the magnetostatic coupling among individual domains plays an essential rôle for the reversal phenomenon of IRM at low temperatures. Even in a single crystal, a small portion of domains may be insensitive to change in magnetocrystalline anisotropy because of local accumulation of internal stress, and the magnetic field of these B domains may affect the other A domains which are magnetically free at the isotropic
temperature. The negative coupling between the A and B domains may result in the reversed magnetization of A domains. The A domains thus remagnetized may be blocked on further cooling because of increasing coercivity.

A question involved in this phenomenon will be why the intensity of oppositely remagnetized A domains can exceed that of the normal magnetization of B domains at the lower temperatures. At the same time, another question may be raised about the recovery of positive magnetization on reheating process. These problems will be theoretically discussed in a separate paper.

6. Some applications of the low-temperature characteristics of magnetic minerals to rock magnetism research.

Since IRM and VRM are believed to be due mostly to domain wall movements which are essentially subject to the magnetocrystalline anisotropy energy, the observed effective demagnetization of these types of remanence by cooling down below the isotropic temperature in non-magnetic space will be justified as a convenient method of “magnetic cleaning” of NRM of rocks. For example, Ozima, Ozima and Nagata (1964) demonstrated that dispersion factor $k=13.0$ of the original NRM of a serpetinite is improved to $k=25.4$ by the low-temperature treatment.

Another application of the low-temperature characteristics of magnetic minerals will be as a convenient method of identifying magnetic minerals in rocks. Fig.8 illustrates an example of such an application. A rock sample is magnetized in a strong field (say, $H=7500Oe$) at the room temperature and again magnetized into the same direction at the liquid nitrogen tem-
perature (-196°C) by the same field. The curve in Fig.8 represents the intensity of magnetization of the sample on re-heating process from -196°C to +20°C in non-magnetic space. The magnetization which sharply decreases from about -140°C on heating represents the low-temperature IRM of magnetite-rich titanomagnetite and the magnetization which begins to be recovered from about -20°C represents the recovery of IRM of hematite-rich ilmeno-hematite; the flat curve between -125°C to -20°C represents the intensity of IRM of very fine grains of either titanomagnetite or ilmeno-hematite which is insensitive to the phase changes.

Some other examples of such an application have been shown by Nagata, Kobayashi and Fuller (1964).

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