Self-reversal PTRM evidence found in Sakurajima volcanic rocks

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Abstract

Self-reversal pTRM (partial thermo-remanent magnetization) evidence was found by direct measurement of pTRM in narrow temperature ranges in Sakurajima volcanic rocks. Self-reversal pTRM was observed at 460°C~490°C in annealed samples of An-ei pumice, and at double ranges of 245°C~260°C and 330°C~340°C in raw samples of An-ei lava and Osumi pumice fall deposit. In the former case, pTRM carrier could not be hemo-ilmenite because it appeared at much higher temperature than the Curie temperature (Tc) of hemo-ilmenite which was reported under 400°C, generally 200°C~300°C.

Partial self-reversal magnetization might be caused by interaction between the two phases of titano-magnetite with different blocking temperatures. Different magnetic mineral phases could be seen in repeated initial susceptibility measurement in ascending and descending temperature with variable maximum temperature. Besides, self-reversal pTRM disappeared after upon the stronger field from 200μT to 550μT according to the samples.

Famous Haruna volcanic rock dominated by hemo-ilmenite was also studied for comparison.

Key words: Self-reversal magnetism, volcanic rocks, Sakurajima, Haruna.

1. Introduction

Self-reversed natural magnetism was first discovered by Uyeda (1958) from the pumice at Haruna volcano in Japan. Recently it was further studied by Ozima and Funaki (2001), Nakai (2002) and Ozima et al. (2003) both on geological and magnet-mineralogical point of view. Beside the Haruna, at Mt. Natib (Kennedy, 1981), Mt. Shasta (Lawson et al., 1987), Nevado de Ruiz (Heller et al., 1986; Haag et al., 1990: Fujiwara et al., 1992), Pinatubo (Ozima et al., 1992; Hoffmann and Fehr, 1996: Bina et al., 1999: Goguitchaichvili and Prevot, 2000) and Mt. Sanpei (Sawada et al.,...
self-reversal behaviors were also found. As to the origin of the self-reversal magnetism, hemo-ilmenite was considered.

General theory of mechanism on self-reversal was summarized in Dunlop and Ozdemir (1997) and McElhinny and McFadden (2000). The mechanism of two phases with different unblocking temperatures was introduced: One phase with the higher unblocking temperature becomes magnetized first, parallel to the external field, and when the magnetic field of the first phase swamps that of the external magnetic field, the second phase with the lower unblocking temperature subsequently becomes magnetized anti-parallel to the first during the cooling. When the magnetism of the lower unblocking temperature is larger than that of the higher, natural self-reversal remanent magnetism can be observed. In this two phase model, hemo-ilmenite is not needed. Only the interacted ferri-magnetic mineral with different blocking temperature is necessary. Sakurajima volcanic rocks were selected in this study because unexpected paleo-geomagnetic intensity data had been found in them (Ueno, 1997).

Detailed pTRM experiment was conducted at intervals of 10℃~50℃ on annealed or raw samples of Sakurajima volcanic rocks. For comparison, Haruna Hutatudake rock was also studied. Yu et al., (2001) reported a reversal remanence observed during high thermal demagnetization (500℃~580℃) in Kurokami pumice of Sakurajima volcano, and they suggested it to be originated by chemical alteration of chromites. However, it was not observed in present study.

2. Samples

The eruptions of the Sakurajima volcano have been recorded in historical documents, and the most recent one is keeping in action since 1955. Products of eruption are all dacitic volcanic rocks. The main eruptions with volcanic rocks are Tenpyohouji eruption (764-766), Bunmei eruption (1471-1476), An-ei eruption (1779-1783), Taisyo eruption (1914-1915) and Syowa eruption (1946). Samples were collected from these eruption sites and Osumi fall deposit dated as 22ka (Kobayashi, 1988). Bunmei pumice (S2-2), An-ei pumice (S2-3) and Taisyo pumice (S2-4) were collected from Nagasakibana quarry. Sample No. S4-2 is a pumice from Nabeyama. At An-ei crater in Kitadake, a pumice (S6-1) was collected. Taisyo pumice (S7-2), An-ei pumice (S7-3) and Bunmei/Kitadake pumice (S7-4) were collected from Amidagawa in Kitadake. An-ei pumice (9621A, 9621B, 9623) was collected in Hurusato town. Dacitic lava which had been used for paleo-intensity measurement (Ueno, 1997) was also studied again. They are Tenpyohouji lava (SCO8) in Kagasikibana, Bunmei lava (SD15) in Kurokami town and An-ei lava (SF17) in Arimura town. Osumi pumice was collected at Nishi in Tarumizu city.
In Fig. 1, sampling sites in Sakurajima and Osumi are shown. From the Haruna volcano, dacitic lava of Hutatudake eruption (1.46 ka) was collected near the Miharasidai stop of cableway at Kaminoyama, Ikaho town. The flow of the lava is flat with 5cm-10cm in thickness, and of two different density parts (Haruna 17-6, 17-7).

3. Characteristics of Natural Remanent Magnetization

Many kinds of experiments were carried in this study, such as alternating field
demagnetization, thermal demagnetization, application of partial thermal magnetism in rough interval of about 30~50 degree, thermal analysis of initial susceptibility, hysteresis, and thermal analysis of saturation magnetism. An-ei pumice (9621A, 9621B, 9623), An-ei lava (SF17), Osumi pumice were applied for the experiment of detailed partial thermo–remanent magnetization. In this section, general characteristics of natural remanent magnetization are described.

**Alternating Field Demagnetization**; In Fig. 2, results for two pumices of Sakurajima (S2–2, 9621B), and Haruna sample are shown. In Fig. 10 and Fig. 13, data for sample No. 9621A and SF17 are also shown.

**Thermal Demagnetization**; Fig. 3 shows the results for two pumices of Sakurajima (S2–2, S2–4) and a Haruna sample. In Fig. 10 and Fig. 13, data for sample No. 9621A and SF17 are also shown.

**Hysteresis**; Fig. 4 shows the results for pumice and lava of Sakurajima. Hysteresis of Osumi pumice and Haruna are also shown. In Fig. 10 and Fig. 13, data for sample No. 9621A and SF17 are also shown.

**Day Diagram** was plotted in Fig.5. Most samples are plotted in multi-domain area.

**Thermal analysis of saturation magnetization**; In Fig. 6, results for pumice of Sakurajima (S2–2, S7–2), and 2 Haruna specimens are shown. In Fig. 10 and Fig. 13, data for sample No. 9621A and SF17 are also shown.
Fig. 2 Alternating field demagnetization (AFD)
measured with AF-demagnetizer and spinner magnetometer made by Natuhara Giken Co.,
(S2-2 4.64 g, 9621B 3.36 g, HARUNA17-6 9.56 g)
Fig. 3 Thermal demagnetization (TMD) measured with the thermal demagnetizer made by Natukura Giken Co.

(S2-2, 30°C, 4.0 mg, 4.5 kg, HARKINA(B), 8.35 E)

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Fig. 4  Hysteresis curves measured with VSM made by Riken Denshi Co. at the maximum field of 10 kG.

Fig. 4-1  Sakurajima pumice
(S2-2 0.17 g, S2-3 0.46 g, S2-4 0.55 g, NABE4-2 0.77 g, S6-1 0.59 g, S7-2 0.12 g, S7-3 0.32 g, S7-4 0.17 g, 9621B 0.50 g, 9623 0.42 g)
Fig. 4-2 Sakurajima lava
(SC08 1.41 g, SD15 0.64 g)

Fig. 4-3 Osumi and Haruna
(OSUMI 0.55 g, HARUNA17 0.62 g)

Fig. 5 Linear plots (Day diagram) and bi-logarithmic plots (Parry plots) of hysteresis parameters.
SD: single domain, PSD: pseudo-SD, MD: multidomain (for titanomagnetite)
Line A: SD+MD mixtures, line B: PSD+MD mixtures, line C: single size (i.e., SD, PSD, MD) (for synthetic magnetite)
4. Application of thermo–remanent magnetization

For the Sakurajima samples, application of partial thermo–remanent magnetization (pTRM) in rough temperature interval (30°C ~ 50°C) was carried out on the sample after pretreatment of alternating field demagnetization or thermo–demagnetization at 700°C. As shown in Fig. 7 and Fig. 8, TRM decreases within some temperature intervals in all the specimens. In the intervals of decreased TRM, self–reversal component might be mostly detectable. Some of the samples were used to apply precise pTRM experiment of narrow interval of about 10°C. Details are described in the next section.
Fig. 7 Application of pTRM in rough temperature interval (after AFD) measured with thermo-demagnetizer made by Natuhara Giken Co.

TRM acquisition curve of Sakurajima pumice after AFD

![Graph showing TRM acquisition curve with different samples labeled: S2-2B, S2-3A, S2-4T, S7-2T, S7-4K.](image)

Temperature (°C)

pTRM acquisition curve of Sakurajima pumice after AFD

![Graph showing pTRM acquisition curve with different samples labeled: S2-2B, S2-3A, S2-4T, S7-2T, S7-4K.](image)

Temperature (°C)

Fig. 7-1 Sakurajima pumice (Laboratory field 50μT)

(S2-2B 2.09 g NRM 6.13×10^{-4} emu AFD800G, S2-3A 2.11 g 3.32×10^{-1} emu 800G, S2-4T 2.66 g 3.00×10^{-1} emu 1000G, S7-2T 3.24 g 5.00×10^{-1} emu 1400G, S7-4K 2.06 g 1.78×10^{-3} emu 1200G)
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Fig. 7-2 Sakurajima lava (SC08, SF17) and Kitadake (S6-1) (Laboratory field 50 μT)

(SC08N: 4.39 g NRM: 7.19E×10⁻⁵ emu, SF17A: 4.84 g 5.10×10⁻⁵ emu, S6-1K: 1.85 g 4.32×10⁻⁵ emu)
TRM acquisition curve of Osumi pumice after AFD 600oe

pTRM acquisition curve of Osumi pumice after AFD 600oe

Fig. 7-3  Osumi pumice (Laboratory field 50μT)
(OSUMI-P  2.09 g  NRM1.95×10^{-4} emu)
Fig. 8 Application of pTRM in rough temperature interval (after 700°C 10min. THD)

![TRM acquisition curve of Sakurajima pumice after 700°C DTH](image1)

![pTRM acquisition curve of Sakurajima pumice after 700°C DTH](image2)

Fig. 8-1 Sakurajima pumice (Laboratory field 100μT)
(S2-2 1.07 g NRM 5.13×10⁻³ emu, S2-3 3.41 g 6.25×10⁻³ emu, S2-4 2.63 g 2.80×10⁻³ emu)
Fig. 8-2  Sakurajima pumice (Laboratory field 100 μT)
(9621A2 3.07 g NRM 1.14×10⁻³ emu, 9621B 2.64 g 8.91×10⁻⁴ emu, 9623 1.83 g 1.04×10⁻⁴ emu)
Fig. 9 shows the TRM result of Haruna Hutatudake volcanic rock. Strong self-reversal component appeared from 220°C~280°C in black part of the sample, corresponding to the Curie temperature of hemo-ilmenite. Intensity of thermo-remanent magnetization (TRM) under certain temperature is similar to the summation of pTRM. Between 380°C and 500°C, pTRM shows normal direction (Fig. 14-3), but TRM shows reversal because the summation of reversed magnetization is larger than that of normal magnetization in these temperatures. Haruna rock used in this study is pumice-like lava of board shape. In red part, TRM shows reversal from 300°C to 600°C. After annealing at 700°C, this attendance becomes stronger. The reason considered is that magnetization of titano-magnetite directed to normal direction becomes weaker after oxidation to hematite.

5. Thermo-remanent magnetization in narrow temperature range

Detailed pTRM experiment was conducted at intervals of 10°C~50°C on annealed or raw samples of An-ei pumice (9621A, 9621B, 9623), An-ei lava (SF17) and Osumi pumice (Osumi). For example, in the case of sample 9621A of An-ei pumice, minimum pTRM could be seen in the ranges of 240°C~270°C and 380°C~450°C (Fig. 11). To clear the reason, detailed pTRM experiment at finer intervals of 10°C~15°C was carried on these minimum pTRM blocks. In present experiment, pTRM was not calculated but directly measured. For instance, pTRM between 250°C and 260°C was measured after heating the sample up to 260°C then cooling in the created laboratory field to 250°C, below 250°C to room temperature the created laboratory field was switched off to zero. The raw sample results as shown in Fig. 11-1, pTRM pointed to normal direction in all temperature intervals. However, after annealing the same specimen at 550°C for 11 hours, then following to 450°C for 60 hours, the same specimen obtained a reversal pTRM between 460°C and 490°C (Fig. 11-2). The finer measurement cleared that the strongest self-reversal magnetization appeared between 475°C and 490°C. If the temperature range was stretched to 430°C~490°C, the pTRM will become normal because normal magnetization part has overcome the reversal one. Samples of 9621B and 9623 showed similar behaviors (Fig. 12).

Contrary to An-ei pumice, An-ei lava and Osumi pumice presented self-reversal pTRM behaviors even in raw samples. Fig. 14 showed the results of raw samples of An-ei lava, Osumi pumice and Haruna pumice. Both samples obtained self-reversal magnetization in narrow range around 250°C and 340°C. Although this kind of precise pTRM experiment was carried on limited number of samples, many examples with decreasing TRM blocks can be observed as shown in Fig. 7 and Fig. 8, in which self-reversal partial magnetization might be found by narrower tem-
Fig. 9 TRM (not pTRM) results of Haruna sample measured with thermo-demagnetizer made by Sogo Kaihatu Co. and JR-5A spinner magnetometer made by AGICO. Laboratory field 25μT for the upper Fig.
Fig. 10  AFD, THD, Hysteresis and Js-T for 9621A.

Fig. 10-1  AFD and THD
(THD 2.07 g NRM 8.03×10⁻⁴ emu)
Fig. 10-2  Hysteresis and Js-T  
(0.184 g  5 kG)

Fig. 11  Narrow temperature pTRM of 9621A raw and 9621A annealed  
measured with thermo-demagnetizer made by Sogo Kaihatu Co. and JR-5A spinner magnetometer  
made by AGICO. Laboratory field 100 μT

Fig. 11-1  9621A raw

Fig. 11-2  9621A annealed
Fig. 12  Narrow temperature pTRM of 9621B and 9623 annealed. Laboratory field 100μT

Fig. 12-1  Annealed 9621B

Fig. 12-2  Annealed 9623

perature analyses.

6. Thermal analysis of initial susceptibility

To see the variation of magnetic phases, thermal analysis of initial susceptibility was measured. The initial susceptibility reflects phase change of magnetic mineral very well. If the distribution of Ti and Fe changes according to the transfer of Ti in titano–magnetite, the initial susceptibility of titano–magnetite must be changed sensitively. The magnetic phases change can be observed by repeated measurement of initial susceptibility in ascending and descending temperature with different maximum temperature.

As shown in Fig. 15, in sample 9621B, a large peak appeared at 470℃ in the first heating of raw sample. However, after annealed, new phases at 334℃ and 580℃ could be observed. Similar attendance can be seen in other samples. In Fig. 16, results are shown about An–ei pumice (9621A, 9621B, 9623), An–ei lava (SF17), Bunmei pumice (S2–2) and Osumi pumice fall (Osumi) applied to repeated mea-
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Fig. 13-2 Hysteresis and Js-T
(0.322 g 5 kG)

Fig. 14 Narrow temperature pTRM of An-ei lava, Osumi and Haruna raw. Laboratory field 100 μT

Fig. 14-1 An-ei lava SF17 raw

Fig. 14-2 Osumi raw
measurements of thermal analysis of initial susceptibility in raw state of samples. Fig. 17 showed results of single run of other samples.

From these experiments of Sakurajima samples, magnetic multi-phases could be found which are supposed to play rolls of A phase and B phase in two phases model of self-reversal. Multi-phases might be appeared because Ti element moves easily in titano-magnetite grains in heating, to create phases of different Curie temperature.

In Haruna sample (Fig.18), a higher phase appeared from 410°C to 530°C, and a weak but stable lower phase (hemo-ilmenite) could be seen at 234°C. Hemo-ilmenite is well known as self-reversal ferri-magnetic mineral.

7. Dependence on applied laboratory field

If the self-reversal magnetization observed in Sakurajima volcanic rocks was due to the interaction of two phases, intensity of applied laboratory field would affect the result greatly. When applied field is strong enough to overcome reversal field created by phase A, phase B will obtain normal magnetization.

In Fig.19, results are shown about dependence of pTRM to applied laboratory field on sample 9621A, 9621B and 9623. As shown in Fig.11, sample 9621A is self-reversal between 465°C and 495°C, and normal in other temperature intervals. PTRM was measured under different laboratory fields in those temperature intervals including self-reversal range. Even in self-reversal temperature range, self-reversal disappeared above 405 μT of laboratory field. The critical field of disappearance changed according to the samples from 190 μT (9623) to 520 μT (9621B). The dependence of self-reversal on applied laboratory field suggests that the two phase model is reasonable in the case of Sakurajima volcanic rocks.
Fig. 15 Thermal analysis of initial susceptibility of 9621B measured with KLY-3S Kappabridge magnetic susceptibility meter made by AGICO. (0.252 g in air, 37μT applied)
Fig. 16 Thermal analysis of initial susceptibility (in air, $377 \mu T$ applied).

Fig. 16-1 9621A (0.212 g), 9621B (0.252 g)
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Fig. 16-2  9623 (0.186 g), S2-2 (0.313 g)
Fig. 16-3  SF17 (0.623 g), Osumi (0.219 g)
Fig. 17 Single measurements of thermal analysis of initial susceptibility (in air, 377 μT applied).
(S2-3 0.211 g, S2-4 0.234 g, S4-2=NABE4-2 0.211 g, S7-2 0.249 g, S7-3 0.219 g, S7-4 0.212 g)
Fig. 18  Thermal analysis of initial susceptibility of Haruna.
(0.336 g in air, 377 μT applied)
Fig. 19  Dependence of pTRM on applied field.

The dependence curve of pTRM to applied field
Sample: 9621A

Fig. 19-1  9621A

The dependence curve of pTRM to applied field
Sample: 9621B

Fig. 19-2  9621B

The dependence curve of pTRM to applied field
Sample: 9623

Fig. 19-3  9623
Fig. 20 Microscopic observation and Bitter pattern of 9621B.

Fig. 21 Bitter pattern experiment of Haruna.
8. Microscopic Observation and Bitter Pattern

Microscopic observations using an optical microscopes as well as domain structure by Bitter techniques were performed to Sakurajima sample 9621B and Haruna volcanic rock. The photos are shown in Fig. 20 and Fig 21. These are preliminary experiments, and differences between the two could not be found. More experiments are needed.

9. Result and Conclusion

Sakurajima volcanic rocks were found to show self-reversal partial thermal remanent magnetization (pTRM) in narrow ranges of temperature. Detailed pTRM experiment at intervals of 10°C~15°C was conducted on raw and annealed samples. Self-reversal pTRM was observed at 460°C~490°C in annealed samples of An-ei pumice, and at double ranges of 245°C~260°C and 330°C~340°C in raw samples of An-ei lava and Osumi pumice. In the former cases, pTRM carrier could not be hemo-ilmenite because it appeared at higher temperature than the Curie temperature (Tc) of hemo-ilmenite which was reported under 400°C, generally 200°C~300°C. The possible mechanism is the model of two phases with different blocking temperatures: One phase with the higher blocking temperature becomes magnetized first, parallel to the external field, and when the magnetic field of the first phase swamps that of the external magnetic field, the second phase with the lower blocking temperature subsequently becomes magnetized anti-parallel to the first. Proceeding experiments supported this two phases model. Thermal initial susceptibility of An-ei pumice changed after annealing from the single phase with Tc of 470°C to three phases with different Tc of 340°C, 470°C and 580°C. Besides, self-reversal pTRM which appeared in the field of under 100μT disappeared after upon the stronger field from 190μT to 520μT according to the samples. Yu et al. (2001) reported a reversal remanence observed during high thermal demagnetization (500°C~580°C) in the samples Kurokami pumice in Sakurajima, and they suggested it to be originated by chemical alteration of chromites. It seems it was not the present case.

Samples used in this study are plotted in pseudo-single domain or multi domain area in Day diagram. Self-reversal in narrow temperature range must be one of the reasons of incorrect paleo-magnetic intensity obtained from pseudo-single domain or multi domain sample.

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References


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要旨

上野直子・鄭重・上野宏共:狭い温度範囲でおきる自己反転残留磁化の検証

岩石が地球磁場中で冷却する際、通常は地球磁場方向の自然残留磁化を獲得する。地球磁場と逆向きの磁場を獲得したとき、自己反転残留磁化と呼ぶ。この現象は自然界で榛名軽石をはじめとして、いくつも報告されている。

自己反転のメカニズムは磁化獲得温度が違う強磁性鉱物 (A, B) が接しているとき、まず高温で磁化を獲得する鉱物 A がある温度で地球磁場方向の磁化を獲得する。温度が下がると低温で磁化を獲得する鉱物 B が磁化を獲得する。このとき鉱物 B がさらされている磁場の向きは、鉱物 A がつくる磁場（反地球磁場方向）と地球磁場の和に向けて、鉱物 A による反磁場が地球磁場よりも大なら鉱物 B は自己反転残留磁化を持つ。鉱物が獲得する磁化の大きさは鉱物の磁化率と外部磁場によるので鉱物 A と鉱物 B がそれぞれ獲得した磁化の大きさよっては岩石全体として自己反転残留磁化をもつことになる。

今まで、榛名軽石をはじめとして自己反転磁化が見られる岩石にはヘモイルメナイトが確認されている例が多いために、自己反転磁化の原因はヘモイルメナイトの存在であると言われてきた。しかし、上記の獲得モデル（2 相モデル）で考えると特定の鉱物が存在する必要はない。磁化獲得温度や磁化率に差のある鉱物が接においていればよい。この考えを検証するために、部分熱残留磁化（pTRM）の変化率に正負がある試料を選び、磁化が減少する温度範囲について狭い温度区分で pTRM を直接測定する実験をおこなった。試料として桜島の溶岩・軽石と大隈軽石を用いた。今回、狭い温度範囲で pTRM 実験を行ったすべての試料について自己反転を獲得する温度範囲が特定できた。また、生試料では自己反転が確認できない試料でも、焼き鈍すと自己反転温度が確認できることができた（Fig. 11, Fig. 12, Fig. 14）。なお、これらの試料はヘモイルメナイトではなく、チタン磁鉄鉱が主として磁化を担っており、さらに、擬似単磁区ないしは多磁区構造をもつつことを飽和磁化の温度変化やヒステリシスパラメーターで確認した。

2 相モデルでは外部磁場が大きい場合は、鉱物 A の磁化によって生じる磁場よりも外部磁場が常に大きくなり、鉱物 B は外部磁化方向の磁化を獲得し、反転磁化を持っていない。すなわち、小さい外部磁場中で自己反転磁化を獲得した岩石でも、大きな外部磁場中では自己反転磁化を獲得しないことを確認した（Fig. 19）。

次に、磁化獲得温度が異なる強磁性鉱物が存在することを確認するために初期帯磁率の温度変化と変化率を調べた。帯磁率の変化率の極大になる温度が、キュリー温度にはほぼ対
応する。今回の試料では温度変化によってキュリー温度が異なる、すなわち磁化獲得温度が異なる相が種々出現することがわかった（Fig.15, Fig.16）。このことは、隣り合う相の磁化獲得温度や磁化率の組み合わせが適当になり、自己反転を獲得する機会があることを示している。

また、反転温度が460℃〜490℃でヘモイルメナイトのキュリー温度以上の高温でしか現れない場合は、ヘモイルメナイトが自己反転を起こしたとは考えにくい。

そこで、比較のためにヘモイルメナイトが自己反転を起こした榛名二つ岳の軽石的溶岩についての実験結果も示した。ヘモイルメナイトのキュリー温度は200℃〜400℃でこの温度近くでヘモイルメナイト自身が自己反転する。岩石全体が自己反転磁化を示すのは、岩石に含まれるその他の強磁性鉱物の磁化がヘモイルメナイトの反転磁化より弱いときである。高温で酸化したチタノマグネタイトなどの磁化がヘモイルメナイトの反転磁化より強くなると反転を示さない（Fig.9）。

さらに、顕微鏡下でピッターパターンによる磁区構造を観察したが（Fig.20, Fig.21）、検討には至っていない。

以上のことから、火山岩は、2相モデルによって条件さえ揃えば容易に自己反転磁化を示しうることが本研究によって明らかにされた。これによって、今まで擬似単磁区ないしは多磁区構造をもつ試料からはしばしば正しい古地球磁場強度が得られないという問題があったが、その原因のひとつとして、狭い温度範囲でおきる自己反転磁化を考慮する必要があることがわかった。