New Observations and Theory of Single-Domain Magnetic Moments

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Abstract. Idealised uniaxial stable single-domain (SD) particles permit only two possible stable positions in which the magnetic moment can lie, either closely parallel or anti-parallel to the particle long (easy) axis. This implicit two state feature has never been challenged in real acicular “uniaxial” SD particles. We now demonstrate that such “uniaxial” particles can allow several quantifiable stable (or metastable) orientations of the magnetic moment within the same particle. A new model is presented with quantitative predictions verified by experiments. The results have important implications for rock magnetism, palaeomagnetism, and magnetic materials research. Firstly, the new model quantitatively accounts for several previously unexplained diverse phenomena exhibited by such SD particles, including the acquisition of gyroremanences, field-impressed anisotropy, and transverse components of remanence in individual particles. These phenomena are theoretically impossible in idealised uniaxial SD particles, and could now be used to quantify the deviation of real particles from ideal behaviour. Secondly, deflections of the natural remanence vector and computations of the ancient field vector and palaeointensity are not only controlled by the shape and distribution of the particles, but also by the possible stable orientations of the moments within single-domain particles. The model is also relevant to other single-domain particle morphologies.

1. Introduction
An idealised uniaxial SD particle has only two possible stable states (figure 1(a)) in which the moment can lie [1]. In nominal “uniaxial” SD particles, such as acicular γ-Fe₂O₃ particles, this implicit two state feature has never been challenged, whilst there has been considerable debate concerning the mechanism of moment reversal between the two states. However, any model of such particles has to explain several observed phenomena, which theoretically should not occur in idealised uniaxial SD particles. These include transverse components of remanence in individual particles [2], field-impressed anisotropy [3-5], and gyroremanences [6-9]. The present paper will demonstrate that a new simple model of “uniaxial” SD magnetic moments can quantitatively explain these diverse phenomena. New predictions are verified by further experiments. The implications of the new model for improving computations of the ancient field vector and palaeointensity in anisotropic rocks will also be briefly discussed.

2. A new model of stable SD moments
A new model is proposed whereby the moment of an acicular (cylindrical) “uniaxial” SD particle has several stable (or metastable) positions. It is further proposed that the range of stable positions is
influenced by the aspect ratio of the particle, being constrained within parallel and anti-parallel cones whose angular dimension $\phi = \tan^{-1} \left( \frac{\text{particle diameter}}{\text{particle length}} \right)$. Figure 1(b) shows a side cross-sectional view (arbitrarily in the xy plane) of the range of parallel ($m_{1a}$ to $m_{1b}$) and anti-parallel ($m_{2a}$ to $m_{2b}$) stable moment positions. There may also be more extreme positions of the moment, especially if strong fields are applied perpendicular to the particle long axis, and also in irregularly shaped particles (those that may not be perfect cylindrical shapes).

![Figure 1](image)

**Figure 1.** (a) Schematic of idealised uniaxial SD particles (prolate ellipsoids). The moment has only two possible opposing stable moment orientations ($m_1$ and $m_2$). (b) Schematic of real “uniaxial” acicular SD particles showing the range of possible “parallel” and “anti-parallel” moment positions ($m_{1a}$ to $m_{1b}$ and $m_{2a}$ to $m_{2b}$) as proposed in the new model. (c) A steep DF (direct field) below the switching field orients the respective moments towards $m_{1a}$ and $m_{2a}$, where they can remain after the field is removed. (d) Above the switching field the DF orients all the moments towards $m_{1a}$.

The model was quantitatively tested by examining the dependence of isothermal remanent magnetisation, IRM, on applied field direction using a sample containing dilutely dispersed (0.03% by volume) acicular SD $\gamma$-Fe$_2$O$_3$ particles (1 $\mu$m in length and 0.22 $\mu$m in diameter). The particles were set in resin in the presence of a direct field (DF) of 100 mT along the x axis. This should ideally have aligned all the particle long axes along x (scanning electron microscopy images on one flat end face of the sample were consistent with this). If the sample comprised ideal uniaxial SD particles fully aligned in the x axis, any subsequent imparted remanence would remain along x regardless of the applied field direction. If the sample comprised ideal uniaxial SD particles partially aligned along x, then the
resulting remanence vector due to any misaligned particles should be coincident with that theoretically calculated using the sample’s IRM anisotropy ellipsoid [10]. If the particles are aligned along x, but the moments have a range of possible stable positions within ± φ as proposed in the new model, then the sample’s resultant remanence direction will be oriented up to ± φ degrees from the x axis for applied fields above the switching field of the particles.

For a relatively steep orientation of the DF the model predicts that as the field strength is increased above the switching field the remanence direction will saturate at the orientation φ, since more and more moments switch from m2a (figure 1(c)) to m1a (figure 1(d)). Figure 2 demonstrates this for a relatively steep DF applied in the xy plane at a declination θF(xy) = +60°. As the applied DF is increased in strength from an initially tumble alternating field (AF) demagnetised state, the declination of remanence, θR(xy), saturates at almost exactly the theoretical value of φ (12.53°) for these particles (aspect ratio 4.5). This is significantly different from the computed declination (28.3°) from the sample’s IRM anisotropy ellipsoid (whose principal axes are oriented along the x, y and z sample axes). Moreover, θR(xy) also saturates at φ when the sample is initially given a strong DF along x (aligning the moments along x) and a DF then applied at increasing strengths at θF(xy) = +60°.

A range of other applied field orientations was tested. Figure 3 shows the experimental results (squares) for a 120 mT DF applied at various declinations, θF(xy), in the xy plane of the sample. Tumble AF demagnetisation was employed prior to each DF application. The results are not consistent with ideal uniaxial SD particles fully aligned along x, except at very small angles of θF(xy). They are also significantly different to the computed values (diamonds) using the sample’s IRM anisotropy ellipsoid [10] at 120 mT, and are thus inconsistent with a distribution of partially aligned ideal SD uniaxial particles. The new model, however, is consistent with the experimental results. For shallow and moderate applied field orientations the declination is no greater than φ. Significantly, there is a distinct plateau at φ at moderately steep angles, which is consistent with the model. At very steep values of
θ_{F(xy)}$, for example the results at θ_{F(xy)} = +75°, the 120 mT applied field may not be above the switching field [11], which may explain why θ_{R(xy)} > φ for this orientation. (Note we believe particle interactions play a negligible role in our results, since the particles are dilutely dispersed and the sample exhibits SD susceptibility versus remanence anisotropy characteristics typical of non-interacting particles. Also, results on isotropic dilute dispersions of SD maghemite and hematite, where the particles were not set in a magnetic field, are also consistent with the new model as detailed later).

![Figure 3](image)

**Figure 3.** The relationship between the applied (120 mT) DF direction in the xy plane, θ_{F(xy)}, and the remanence direction, θ_{R(xy)}, for a dilute dispersion of SD γ-Fe₂O₃ particles (aspect ratio 4.5; φ = 12.53°) aligned along the x axis. Errors are smaller than the symbol size. Squares are experimental results. Diamonds are computed values [10] using the field directions and the sample’s IRM anisotropy ellipsoid at 120 mT. Ideal uniaxial SD particles fully aligned in the x axis would have plotted solely along the x axis.

### 3. Explanation for field-impressed anisotropy effects in SD particles

Theoretically, a strong AF or DF applied to an ideal uniaxial SD particle should not cause its low field susceptibility anisotropy to change, since the two possible states shown in figure 1(a) are effectively identical as regards reversible susceptibility in a weak AF. By a similar argument a distribution of such particles should not exhibit any difference in susceptibility between the demagnetised and magnetised states. Therefore, observations of field-impressed susceptibility anisotropy in dilutely dispersed “uniaxial” SD particles from magnetic tape [3,4] and rocks [5] require an explanation. However, if SD particles have a range of stable moment positions then these effects can be explained. For example, after tumble AF demagnetisation the moment of an acicular SD particle could be orientated within the range of possible stable positions shown in figure 1(b). When a strong DF is subsequently applied parallel to the particle long (x) axis the moment will re-align along this axis and remain there once the field is removed. This will change the particle’s susceptibility, unless the moment was already in this orientation, resulting in a minimum susceptibility parallel to x. (Note that an ideal uniaxial SD particle has zero susceptibility parallel to its long axis [10]). Likewise for a random distribution of acicular SD particles, our model predicts that the applied field impresses a susceptibility anisotropy represented by an ellipsoid of revolution with the unique minimum axis in the
field axis, which is consistent with observations [3-5]. We now further predict that particles with lower values of $\phi$ (higher aspect ratios) should exhibit weaker field-impressed anisotropy, since more needle-like particles should be closer to ideal uniaxial behaviour. We tested this in two samples containing randomly distributed (isotropic) and dilutely dispersed (0.03% by volume) particles of SD $\gamma$-Fe$_2$O$_3$, with each sample containing particles of a different aspect ratio. In one sample the particle aspect ratio was 6.5 ($\phi = 8.75^\circ$) and in the other it was 4.5 ($\phi = 12.53^\circ$). The application of an 80 mT DF caused percentage field-impressed susceptibility anisotropies of $-1.22\%$ and $-1.73\%$ respectively, where the negative sign indicates a decrease in susceptibility in the field direction and field-impressed anisotropy is defined by equation 7 of [4]. The impressed anisotropy values were directly proportional to $\phi$ (inversely proportional to particle aspect ratio) confirming our predictions (table 1).

The model was also extended to SD hematite ($\alpha$-Fe$_2$O$_3$). Here, at room temperature, the moment can lie in one of 3 possible easy axes, each separated by 60°, in the hexagonal basal plane. With these three easy axes, then effectively $\phi = 60^\circ$ with three possible “parallel” and three “anti-parallel” stable moment positions. We would therefore predict that SD hematite should exhibit a relatively high field-impressed anisotropy. The application of a DF of 80 mT to randomly and dilutely dispersed SD hematite particles in specular form did indeed result in a high percentage impressed susceptibility anisotropy of $-8.93\%$. Significantly, the result was in virtually the same direct proportion to $\phi$ as for the SD $\gamma$-Fe$_2$O$_3$ samples (table 1).

Table 1. Correlations between $\phi$, field-impressed anisotropy (after a DF of 80 mT) and RRM (at 95 r.p.s., in an AF of 80 mT).

| Particle Type       | $\phi$ (Degrees) | Field-Impressed Anisotropy $A_{fs}$ (%) | RRM ($10^3$ Am$^2$ kg$^{-1}$) |
|---------------------|------------------|----------------------------------------|--------------------------------|
| Idealised Uniaxial SD | 0                | 0                                      | 0                              |
| SD $\gamma$-Fe$_2$O$_3$ (Aspect ratio 6.5) | 8.75             | -1.22                                  | 226                            |
| SD $\gamma$-Fe$_2$O$_3$ (Aspect ratio 4.5) | 12.53            | -1.73                                  | 335                            |
| SD $\alpha$-Fe$_2$O$_3$ (Equant platelets) | 60.00            | -8.93                                  | N/A                            |

4. Explanation for gyroremances in acicular SD particles

The new model explains how gyroremences [6-9], rotational remanent magnetisation (RRM) and gyroremenant magnetisation (GRM), can be acquired even in a prolate cylindrical ellipsoid of revolution (gyroremences are theoretically impossible if such a particle behaved like an ideal uniaxial SD particle [8]), and perpendicular to the AF axis. The range of stable moment positions allows the moment to stick in an orientation inclined to the particle long axis, resulting from the transverse bias produced by the gyromagnetic effect. The transverse component of magnetisation so produced (perpendicular to the AF axis) is the gyroremancence. Furthermore, the model predicts that more needle-like SD particles (closer to ideal uniaxial) might acquire a lower gyroremancence. This was tested by measuring the RRM acquired by the two isotropic samples of SD $\gamma$-Fe$_2$O$_3$ particles. The RRM was acquired at a rotation rate of 95 r.p.s. in an AF of 80 mT using a rotational magnetiser. The magnitude of the RRM was directly proportional to $\phi$ (table 1). The sample containing the higher
aspect ratio particles acquired the lower RRM as predicted. (Note that gyroremanences should not theoretically be acquired by a SD particle where the moment is restricted to one plane [8], such as for hematite at room temperature, which is why there are no results for hematite in table 1).

5. Implications for correcting palaeomagnetic deflections in anisotropic rocks
Computations of the ancient field direction and palaeointensity from the deflected natural remanent magnetisation (NRM) recorded in anisotropic rocks normally assume that the deflections are only controlled by the shape and degree of alignment of the particles, and their orientation with respect to the applied field. However, they will also depend on the range of stable moment positions a SD particle can possess from acquiring a remanence (an NRM or, more significantly, a laboratory analogue remanence used to compute the ancient field vector).

Interestingly, the new model permits smaller remanence deflections than those expected from the SD particle alignments, because the angle between the moment and the field direction, $\delta = \theta_{F(xy)} - \phi$ (figure 1(d)), can be less than the angle between the particle long axis and the field direction, $\theta_{F(xy)}$.

6. Conclusions
A new model relates the range of possible stable magnetic moment positions in a SD particle to quantifiable predictions of measurable bulk properties (susceptibility and remanence) in large distributions of such particles. The model provides quantitative explanations for field-impressed effects, gyroremanences, and transverse components of remanence in acicular SD particles, all of which are theoretically impossible in ideal uniaxial SD particles. The deviation from ideal behaviour can be quantified using these effects. New idealised particles (for media storage etc) could be designed and tested using these effects. Also, quantifying the range of stable moment positions within SD particles due to remanence acquisition will improve computations of the ancient field direction and palaeointensity from the NRM recorded in anisotropic rocks.

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