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Magnetostatic Energy of Spherical Two-Domain Particles and the Upper Single-Domain Particle Size in Magnetite

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Abstract. Calculations have been made of the magnetostatic energy of a spherical particle divided by a single 180° domain wall. The results suggest that for magnetite there may be no jump from a uniformly magnetized state to a two domain configuration at a particular particle size, but that the uniform magnetization is preferred for particles $\lesssim 60$ nm in diameter, and distinct domains may appear in particles with diameters $\gtrsim 150$ nm.

Key words: Rock magnetism – Single domain particles – Magnetic domains – Magnetite.

The presence of sub-microscopic magnetic particles in rocks (e.g. Evans and Wayman, 1970, 1974), makes a knowledge of the single-domain size-range, and the properties of particles just larger than single domains, important for palaeomagnetism. Butler and Banerjee (1975) applied Amar's model (Amar 1957, 1958a, 1958b) of a two-domain particle to rectangular parallelepipeds of magnetite, and concluded that the upper limit to the single-domain size-range for magnetite is set by the transition to a two domain configuration with a single 180° domain wall. In Amar's calculation of the magnetostatic energy of the two-domain system, the wall is treated as a uniformly magnetized slab of thickness equal to the effective width of the domain wall, and having the same intensity of magnetization as the neighbouring domains. This overestimates the magnetostatic energy of the wall.

A variation of the calculation, with a different estimate of the magnetostatic energy, is applied here to spherical magnetite particles. Butler and Banerjee preferred the form of a parallelepiped as being more realistic for naturally occuring grains. It is however interesting to see the effect of the change from cubic to spherical shape, while synthetic particles produced by milling may indeed be closer to spherical than cubic form.

Consider a 180° wall of width $2x$ symmetrically dividing a particle of radius R. The volume occupied by the wall is then $2\pi(R^2x - x^3/3)$. Following Amar (1957) the wall energy per unit area (σ) is given by $(\sigma_0/2)(2x/\delta_0 + \delta_0/2x)$ where σ_0 and δ_0 are the wall energy per unit area and the wall width in bulk material. As a first approximation a wall energy per unit volume ($\sigma/2x$) and a total wall energy (E_w) may be calculated.

$$E_w \approx \frac{\pi}{x}\left\{R^2x - \frac{x^3}{3}\right\}\left\{\frac{\sigma_0}{2}\right\}\left\{\frac{2x}{\delta_0} + \frac{\delta_0}{2x}\right\}.$$

The values of σ_0 and δ_0 used by Butler and Banerjee (1975) have been taken: $\sigma_0 = 0.8 \times 10^{-3}\,\mathrm{Jm}^{-2}$, $\delta_0 = 150$ nm.

In the case where the domain wall is considered as having the same intensity of magnetization as the two domains, the magnetostatic problem can be simplified by imagining the wall magnetization rotated through 90° in the plane of the wall so as to be parallel to the magnetization of one of the domains. As the interactions with the two domains are equal and opposite this does not change the magnetostatic energy, and the problem becomes that treated by Néel (1944). Figure 2, curve A, shows the reduced magnetostatic energy (magnetostatic energy of the particular configuration divided by that of the same particle uniformly magnetized) versus reduced wall thickness (x/R) obtained from Néel's solution.

An improved model for calculating the magnetostatic energy is shown in Fig. 1. The wall is divided in halves, in each of which the magnetization is represented by two components of intensity $2/\pi$ times the saturation magnetization. These two components are parallel and perpendicular, respectively, to the magnetization of the neighbouring domains.

Expanding the resulting surface charge distribution in terms of Legendre polynomials (the method used by Néel, 1944 and described by Carey, 1971), and satisfying the boundary conditions at the surface of the sphere for the internal and external potentials, leads to a series solution for the magnetostatic energy E_m. Carey's paper describes the procedure in detail. An important point is that because there is no net interaction between the mutually perpendicular components of magnetization, the energy can be calculated as the sum of two terms, one due to the components parallel and antiparallel to the domain magnetizations, and one due to the net magnetization of the wall.

The magnetostatic energy is given by

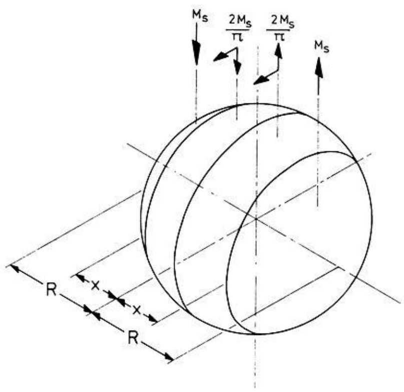

Fig. 1. Spherical particle divided by à 180° domain wall. In the two domains the magnetizations are $+M_s$ and $-M_s$. The rotation of the magnetization in the wall is approximated by giving each half a mean magnetization $2/\pi\ M_s$ parallel to that in the neighbouring domain

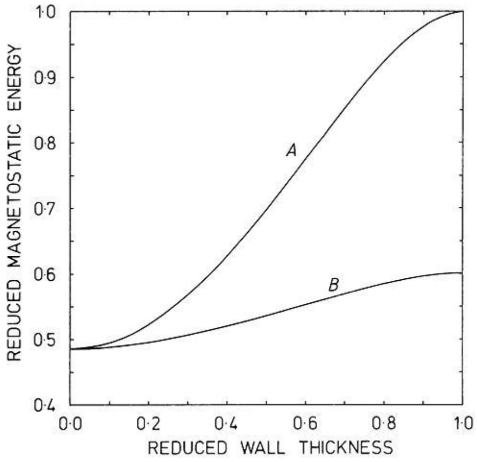

Fig. 2. Reduced magnetostatic energy (magnetostatic energy divided by the magnetostatic energy of a uniformly magnetized sphere of the same size) versus reduced wall thickness (wall thickness divided by diameter of sphere). Curve *A* is for a domain wall carrying magnetization M_s perpendicular to domain magnetizations. Curve *B* refers to the magnetization scheme in Fig. 1. (The energies were calculated to 35 terms in the series expansions)

$$E_m = \tfrac{8}{9}\pi^2 M_s^2 R^3 E_r \qquad \text{(c.g.s.)}$$
$$= \tfrac{2}{9}\pi\,\mu_0 M_s^2 R^3 E_r \qquad \text{(S.I.)}$$

where R is the radius of the sphere, M_s the spontaneous magnetization within the domains, and E_r the reduced magnetostatic energy given by

$$E_r = \frac{\gamma^2}{\pi^2}(3-\gamma^2)^2 + \frac{9}{8}\sum_{n=2}^{\infty}\frac{n(n+1)}{(2n+1)^2}\{a_n^2+b_n^2\},$$

$$a_n = \left(\frac{2}{\pi}-1\right)(1+(-1)^n) \cdot \left\{\frac{P_n(\gamma)-P_{n-2}(\gamma)}{2n-1} - \frac{P_{n+2}(\gamma)-P_n(\gamma)}{2n+3}\right\} - \frac{4}{\pi}\left\{\frac{P_n(0)-P_{n-2}(0)}{2n-1} - \frac{P_{n+2}(0)-P_n(0)}{2n+3}\right\},$$

$$b_n = \frac{2}{\pi}((-1)^n-1)\left\{\frac{P_n(\gamma)-P_{n-2}(\gamma)}{2n-1} - \frac{P_{n+2}(\gamma)-P_n(\gamma)}{2n+3}\right\},$$

$$\gamma = x/R.$$

E_r is plotted as a function of γ in Fig. 2, curve *B*.

Summing the wall and magnetostatic energies and then dividing by the magnetostatic energy of a single domain particle of the same size gives the total reduced energy, which is plotted as a function of wall thickness for various particle diameters in Figs. 3 and 4. The results of the simpler model of the wall magnetization are shown in Fig. 3, while Fig. 4 refers to the model of Fig. 1 and the above equation for E_m. From Fig. 3 it is seen that for particles greater than 113 nm there is a

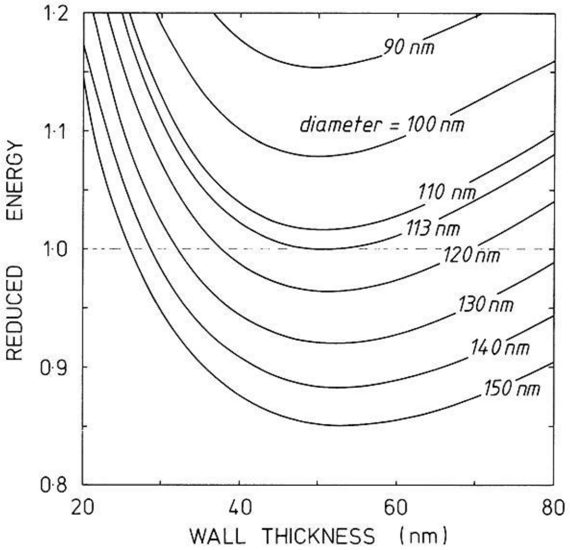

Fig. 3. Reduced energy versus wall thickness for two-domain magnetite particles of various diameters. The magnetostatic energy is taken from Curve *A* of Fig. 2

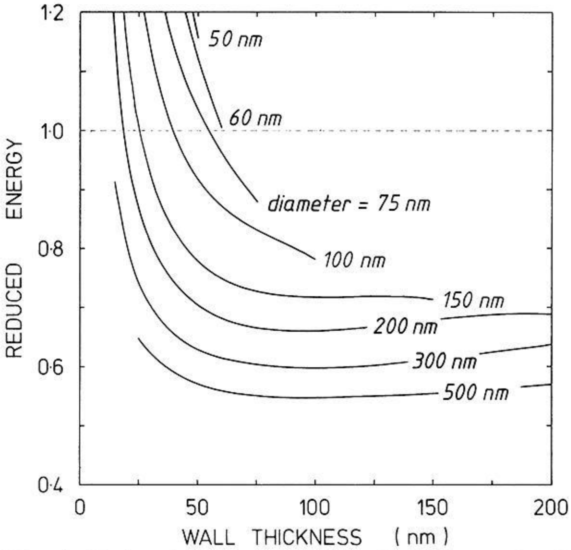

Fig. 4. Reduced energy versus wall thickness for 2-domain magnetite particles of diameters 50 nm to 500 nm. The magnetostatic energy is that of Curve *B*, Fig. 2

domain wall thickness for which the total reduced energy is less than one, and the two-domain state is stable. Taking the ratio of wall area to particle volume as the critical parameter, this transition size agrees exactly with the value 76 nm obtained by Butler and Banerjee (1975) when they carried out the equivalent calculation for cubic particles. Quite a different picture appears in Fig. 4 however. Here the single domain state is energetically favoured for particles smaller than 60 nm, but two distinct domains do not appear until the particles are larger than 150 nm, intermediate sized particles being filled by the wall itself.

Although the above calculations are not rigorous they do suggest that the upper size limit to single domain behaviour in equidimensional magnetite may not be a direct transition to a two-domain configuration, but rather to a domain wall like magnetization as proposed by Dunlop (1973).

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