Effect of the Interaction between Magnetic Particles on the Critical Single-Domain Size

A. H. MORRISH AND L. A. K. WATT
University of Minnesota, Minneapolis, Minnesota
(Received November 13, 1956)

On theoretical grounds, the critical single-domain size of a ferromagnetic particle is expected to be influenced by the interaction of other neighboring particles. Measurements of the coercive force of some γ-Fe_2O_3 powders containing particles of different sizes and shapes at various concentrations provide experimental evidence for the existence of such an effect.

INTRODUCTION

THE critical single-domain size of ferromagnetic particles is usually calculated[1] by considering an isolated particle. The calculation proceeds by comparing the free energy for the state of uniform magnetization with that for a state of nonuniform magnetization. It is found that there is some critical size for the particle below which the single-domain configuration has the lower energy.

Experimentally one is almost never concerned with a single isolated particle but rather with a large number of particles in the form of a powder or a suspension. The interaction between the particles introduces an additional energy term which may be expected to influence the critical size. This communication presents the first experimental evidence for the existence of such an influence.

Kondorskii[2] has made a theoretical investigation of this effect. It is found that for an isolated ellipsoidal particle the critical single-domain size depends on the polar and equatorial demagnetization factors of the particle. For a particle in a powder the demagnetization factors of an isolated particle may be replaced by effective demagnetization factors which take approximate account of the magnetic interactions with neighboring particles. Néel[1] found a relationship of the form

$$D=D_0(1-v) \tag{1}$$

for all values of v, where D is the effective demagnetization factor for a particle in the powder, D_0 is the demagnetization factor for an isolated particle, and $v=d/d_0$ is the powder's volume concentration, d being the powder density and d_0 the density of the bulk material. According to Kondorskii, Néel does not take sufficient account of the effect of particles very close to the one considered. Kondorskii's calculation for elongated particles with random orientation gives the result

$$\begin{aligned} D&=D_0(1-k_0v) \quad \text{for} \quad v<1/k_0,\\ D&=0 \quad\quad\quad\quad\;\; \text{for} \quad v\geqslant 1/k_0 \end{aligned} \tag{2}$$

with $k_0>3$. For ellipsoids of revolution,

$$k_0=\frac{4\pi}{3}\left(\frac{2D_a+D_b}{D_aD_b}\right), \tag{3}$$

where D_a is the demagnetization factor along the polar axis and D_b is the demagnetization factor along an equatorial axis, for an isolated particle.

These results lead to the following conclusions.

(1) If the particles are single domains when isolated, they will remain so at all concentrations.

(2) If the particles are multidomained but are close to the critical size, they may become single domains as the concentration is increased.

Wohlfarth[3] has investigated the effect of the interaction between particles in a more rigorous manner than Kondorskii, but unfortunately his results apply only to certain special cases and cannot be readily compared with experiment.

The basis of our experiment is the measurement of coercive force as a function of volume concentration for various powders whose particles have different mean sizes and shapes. For a powder containing ellipsoidal single-domain particles, the various theories lead to formulas of the form

$$H_c=H_{c0}(1-k_0v-k_1v^{5/3}+\cdots), \tag{4}$$

where H_{c0} is the coercive force at infinite dilution. According to Néel, $k_0=1$, $k_1=0$. According to Kondorskii, k_0 is given by Eq. (3) and $k_1=0$ and the equation is valid only for $v<1/k_0$. On the other hand, Wohlfarth finds $H_{c0}k_0=I_0A$ and $H_{c0}k_1=I_0B$, where I_0 is the saturation magnetization and A and B are constants that depend on the interactions and geometrical arrangement of the particles.

Multidomain particles are expected to show little or no change of coercive force with packing. Morrish and Yu[1] have obtained experimental evidence that confirms the predicted behavior of the coercive force for these two extreme cases.

EXPERIMENT

Four different samples of γ-Fe_2O_3 powders were investigated. The mean particle size and shape varied from sample to sample. The powders were prepared

[1] C. Kittel, Phys. Rev. 70, 965 (1946); L. Néel, Compt. rend. 224, 1488 and 1550 (1947); E. C. Stoner and E. P. Wohlfarth, Trans. Roy. Soc. (London) A240, 599 (1948); A. H. Morrish and S. P. Yu, J. Appl. Phys. 26, 1049 (1955); and W. F. Brown, Jr. Phys. Rev. 105, 1479 (1957).

[2] E. Kondorskii, Izvest. Akad. Nauk. S. S. S. R. Ser. Fiz. 16, 398 (1952).

[3] E. P. Wohlfarth, Proc. Roy. Soc. (London) A232, 208 (1955).

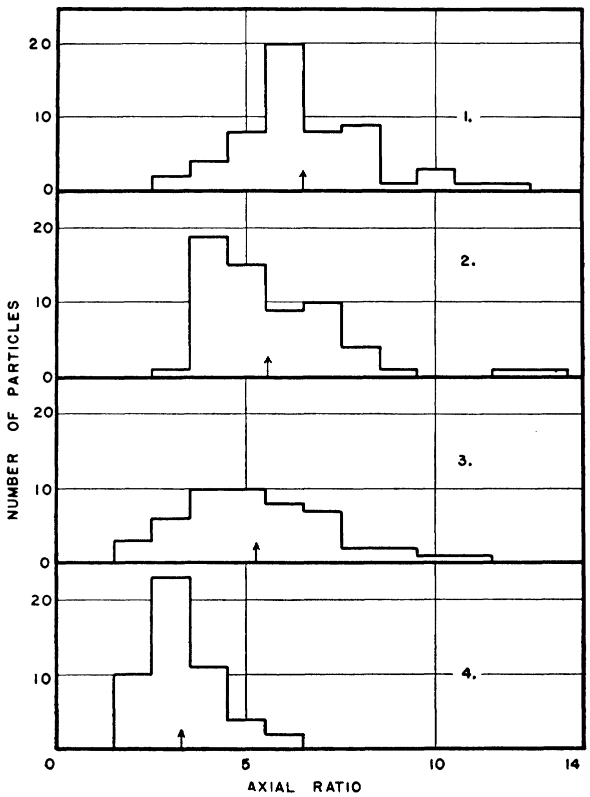

FIG. 1. Histograms of the particle shape distribution in four ferrite powders.

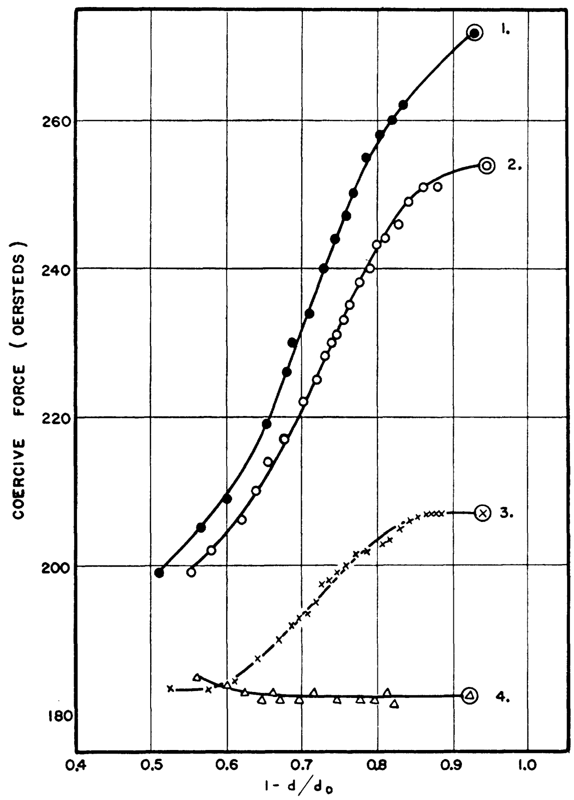

FIG. 2. The coercive force as a function of concentration for four ferrite powders containing different particle sizes and shapes. d=powder density; d_0=density of bulk material.

from the yellow pigment oxide FeO·OH by a cycle of dehydration, reduction, and oxidation in a manner described previously by Osmond.[4] The final product was 99% γ-Fe_2O_3, the impurity probably being mainly water. The pigment oxide particles were grown so that, by controlling the time of growth, samples were obtained having different mean particle volumes. The pigment particles were acicular and the γ-Fe_2O_3 particles retained this shape.

The shape distributions for powders 1 to 4 (Fig. 1) were determined from a series of electron microscope pictures. Measurements were made on about 50 particles of each powder. The axial ratio plotted in Fig. 1 was the ratio of the polar or long axis to the equatorial or short axis, the values being rounded off to the nearest integer. The figure also shows the mean axial ratio for each sample. This ratio ranged from 6.5/1 for sample No. 1, the most acicular, to 3.3/1 for sample No. 4, the least acicular.

The distribution in length of the particles was also determined for each sample. It was found that the lengths ranged from 0.1 to 1.5 microns with a mean somewhere between 0.5 and 0.6 micron for each sample. Thus, the samples did not differ greatly in the lengths of the particles, only in the shape factor.

In analyzing the electron microscope pictures it was found that the particles tended to coalesce into large agglomerates and that this tendency increased as the particle size increased. Thus, a bias may have been introduced in the statistical samples. Further, the number of particles measured in each sample is not sufficient to give the size and shape distribution with great accuracy. However, there is little doubt that the mean axial ratio decreases as we proceed from sample No. 1 to sample No. 4. Measurements of the settling time of the powders were in reasonable agreement with the above data.

The coercive force, the value of the field for which the magnetization is zero, was measured by a method described by Morrish and Yu.[1] The concentration of the sample was increased by compressing the powder in a die. Typical results[5] for the four samples are shown in Fig. 2. For low concentrations there was a possibility that the measured coercive force was lower than the true value because of physical motions of the powder particles under the action of the reverse magnetic field. To test this possibility, the powders were immobilized in paraffin. The coercive force of these very

[4] W. P. Osmond, Proc. Phys. Soc. (London) **B65**, 122 (1952).

[5] The results for sample No. 1 have been published previously by Morrish and Yu. See reference 1.

dilute samples is shown by the encircled points in Fig. 2. The evidence is therefore very strong that the measured coercive force was the true one even for the points of low concentration.

DISCUSSION

Comparison of the mean size and shape of the particles of the four samples with theoretical calculations of the critical single-domain size for an isolated particle leads to the following conclusions. At great dilution, most or all of the particles of sample No. 1 are single domains. On the other hand, most or all of the particles of sample No. 4 are multidomained. Samples Nos. 2 and 3 contain particles close to the critical size.

The experimental decrease in coercive force with compression for sample No. 1 and the constancy of the coercive force for sample No. 4, as shown in Fig. 2, lend support to the above conclusion. Sample No. 2 also seems to consist mainly of single-domain particles although the behavior at low concentrations might indicate the presence of a small percentage of particles above the critical size. Sample No. 3, on the other hand, behaved as though all or most of the particles are multidomained until a value of $1-d/d_0=0.85$ is reached when the coercive force begins to decrease with further packing. This change in the behavior continues until the relationship becomes almost linear, indicating that most of the particles have become single domains. The critical single-domain size has therefore been increased by the compression in agreement with the prediction of Kondorskii.[2] The gradual change is explained by the fact that the sample contains a distribution of particle sizes and shapes.

Kondorskii also suggested that the coercive force might increase as the transition point is approached. Our results show no such increase. However, this may be due to the distribution in particle size and shape in our powders.

The curves for samples Nos. 1 to 3 also show a departure from the linear decrease in H_c with compression at high powder densities. According to Kondorskii there should be a departure from linearity at $v=1/k_0$, where k_0 is given by Eq. (3). If $a/b=6$ (approximately the mean value for sample No. 1), $k_0=8$ and the critical value of $v\approx 0.88$. This large value of v is obviously in disagreement with the experimental results. It is possible that, with an improved model and fewer approximations, Kondorskii's theory could account for the observed behavior.

In addition, an increase in H_c with compression could be caused by (1) the introduction of strains in the particles and (2) breakage of the particles. The fact that sample No. 4 also shows an increase in H_c would seem to support these hypotheses. A further complicating factor is the possibility of bridge formation between the particles, with the subsequent formation of Bloch walls.[6] This effect would decrease the coercive force. It is not possible at this time to decide which of these mechanisms are operative.

CONCLUSION

The experimental results show that the critical single-domain size of acicular magnetic particles contained in an assembly of particles depends on the concentration. The interaction between particles is found to increase the critical size, in agreement with a theoretical prediction of Kondorskii.

ACKNOWLEDGMENTS

We are indebted to A. H. Persoon of the Minnesota Mining and Manufacturing Company for preparing the powders and to J. F. Hartmann and his staff of the University's Anatomy Department for their assistance with the electron microscope work. We wish to thank W. F. Brown, Jr., for stimulating discussions relating to Kondorskii's theory. This research was financed by the Mines Experiment Station.

[6] L. Weil, Revs. Modern Phys. **25**, 324 (1953).