

represent those of an ideal superconducting surface. Nevertheless, this type of experiment seems capable of giving information about the electronic states in a superconductor.

The technique of measuring currents from approximately 10^{-20} ampere to 10^{-17} ampere with a charged particle counter extends the range of measurements

in many electron emission experiments. For instance, studies of weak thermionic emission and weak field emission are two examples of experiments which can be done with this method. Various counters could be employed; Geiger counters, scintillation counters, or electron multiplier assemblies. Such a program is now being set up.

Magnetic Measurements on Individual Microscopic Ferrite Particles Near the Single-Domain Size

A. H. MORRISH AND S. P. YU
University of Minnesota, Minneapolis, Minnesota
 (Received December 27, 1955)

A novel quartz-fiber torsion balance is employed to make magnetic measurements on individual microscopic ferrite particles. A very direct experimental test provides strong evidence for the existence of single-domain magnetic particles. Further, the results confirm theoretical calculations of the critical single-domain size within a small factor.

INTRODUCTION

ON theoretical grounds, there is strong reason for believing that a sufficiently small particle of ferromagnetic material will exist as a single domain. This configuration will result if the associated magnetostatic energy is less than the energy required to form either Bloch walls or else a circular arrangement of the atomic spins. Either of the latter configurations would reduce or remove the magnetostatic energy.

The experimental evidence for the existence of single-domain particles has been obtained by measurements made on powders or suspensions containing many small particles. The evidence presented so far has been at best indirect, and further, can usually be criticized on some additional grounds.

It was thought that an experiment in which an individual microscopic magnetic particle was investigated might provide very direct experimental evidence on whether or not single-domain particles exist. Measurements on an individual particle eliminate several of the factors present in powders. Chief among these are the interparticle interactions and the averaging effect due to the different orientations of the powder particles. Therefore, a novel quartz-fiber torsion balance was constructed.¹ This balance permitted measurements to be made on an individual micron-sized magnetic particle. The relative remanent magnetization of a particle was determined by noting the deflection of the balance in some inhomogeneous magnetic field.

The idea underlying the experiment is as follows. A single-domain particle will have only two, or some small multiple of two, directions along which the magneti-

zation vector will lie. Therefore, the deflection, in the inhomogeneous field, will only be able to assume certain constant values that differ from each other by some relatively large discrete amount. On the other hand, with a multidomain particle, one would expect that the net remanent magnetization, and therefore also the deflection, could be varied in a continuous fashion, up to of course, some maximum amount.

This idea is closely related to the fundamental properties of single-domain particles investigated theoretically by Stoner and Wohlfarth.²

PREVIOUS EXPERIMENTS

It is pertinent to consider the previous experimental evidence for the existence of single-domain particles.

Frequently it is supposed³ that the large coercive force observed for powders containing small particles is evidence for single-domain behavior. This is not conclusive for a variety of reasons. The coercive force increases as the particle size decreases, even in the multidomain region. No evidence has been put forward that there is a sudden discontinuity in the coercive force as the critical size is reached. In fact, there is some evidence that the coercive force continues to increase for particles smaller than the critical size. Further, the absolute value of the coercive force obtained for supposedly single-domain powders is usually considerably less than that predicted by theory.

Another experiment has been carried out by Kittel

² E. C. Stoner and E. P. Wohlfarth, *Trans. Roy. Soc. (London)* **A240**, 599 (1948).

³ F. Bertaut, *Compt. rend.* **229**, 416 (1949); C. Guillaud, thesis, Strasbourg, 1943 (unpublished); W. H. Meiklejohn, *Revs. Modern Phys.* **25**, 302 (1953).

¹ S. P. Yu and A. H. Morrish, *Rev. Sci. Instr.* **27**, 9 (1956).

*et al.*⁴ to determine whether or not a powder was single-domained. The idea of this experiment is that a powder consisting of small single-domain spherical particles (nickel) would require a field of $\approx 2K/I$ for saturation, whereas a powder of larger multidomain nickel particles would require a field of $\approx 4\pi I/3$ for saturation. This second field is much larger than the first; experimental results gave saturation fields of about the predicted amounts. However, as Kittel *et al.* themselves point out, the results show only that the small particles are close to the single-domain size where the exchange energy approaches the demagnetization energy. Further, this does not necessarily mean that slightly smaller particles are certain to be single domained. One must admit the possibility that the size at which the particles become paramagnetic will be reached first, so that no region in which the particles are single domained would exist.

Elmore⁵ measured the magnetization curve of colloidal magnetite and colloidal siderac. If the particles were small permanent magnets, one would expect them to behave as the molecules of a classical paramagnetic gas, and have a magnetization curve given by the Langevin equation:

$$I = I_s(\coth a - 1/a), \quad a = \mu H/kT.$$

Experiment showed that this was so. It was concluded that the colloidal particles were single domained. However, it is possible that small particles have a remanent magnetization, even though they are multidomained. If domain wall motion was small, results of the sort Elmore observed would have been expected. The highest field strength used was 500 oersteds.

Finally, experiments in which powders are compressed

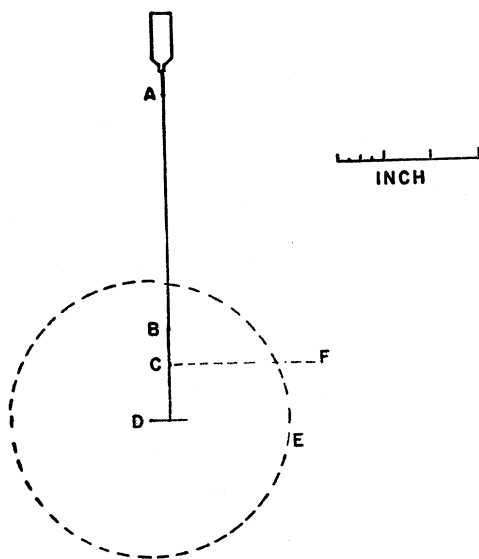


FIG. 1. The suspension system of the torsion balance.

⁴ Kittel, Galt, and Campbell, *Phys. Rev.* **25**, 302 (1950).

⁵ W. C. Elmore, *Phys. Rev.* **54**, 1092 (1938).

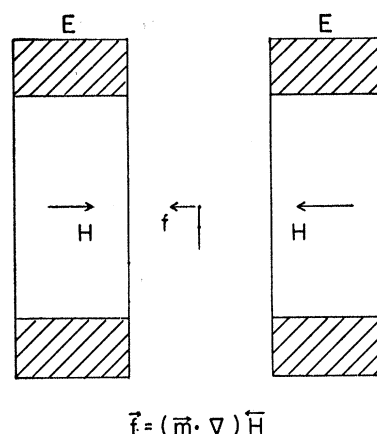


FIG. 2. The balance as seen in a horizontal plane through the "T." H is the magnetic field produced by the solenoids E , connected in opposition, to produce the force on the particle.

may show a decrease of the coercive force as the density of the powder is increased.^{6,7} However, this behavior is expected only for single-domain particles with a shape anisotropy.

TECHNIQUE

A description of the quartz-fiber torsion balance has been published elsewhere.¹ For completeness, a very brief summary will be given here. In order to get a reasonably large deflection with a 1μ particle, the quartz suspension fiber ($A-B$ of Fig. 1) is made very fine, about 0.5μ in diameter. Larger fibers are often used when experiments are performed on larger magnetic particles. Two thicker quartz fibers, forming an inverted "T," hang at the bottom of the fine fiber at B . A mirror cemented to the vertical member of the "T" at C , together with a galvanometer lamp and scale (F), permits observation of a deflection of the system. The magnetic particle is glued at one end of the horizontal quartz fiber (D). The suspension system is housed in a specially designed apparatus (not shown), which overcomes the problem of zero drift. The magnetic particle is magnetized by discharging a bank of condensers charged to some definite voltage through two air solenoids connected in series (E). The deflection for a particular amount of remanent magnetization of the particle is obtained by applying an inhomogeneous field, whose gradient is fairly large, but whose absolute magnitude is small, at least compared to the magnetizing field. The force on the particle is indicated in Fig. 2, which is a drawing of the system in a horizontal plane passing through the lower arm of the inverted "T."

The sequence of measurements made depended on whether the magnetic particle on the balance appeared to behave as a single-domain or a multidomain particle.

If it appeared to be single domained, first a large

⁶ L. Weil, *J. phys. radium* **12**, 437 (1951).

⁷ A. H. Morrish and S. P. Yu, *J. Appl. Phys.* **26**, 1049 (1955).

field pulse, of about 2000 oersteds, was applied in one direction, which for convenience we shall call the negative or left direction. The deflection of the system in an inhomogeneous field was then observed for a given current. The current chosen was such as to give a convenient deflection. Then, a small pulse, of say 100 oersteds, was applied in the reverse, or right (positive) direction. Again the deflection in the same inhomogeneous field was observed. Next, the 2000-oersted field was applied in the left direction, followed by another pulse, of say 200 oersteds, in the right direction. Again, with the same inhomogeneous field, the deflection was observed. The sequence was repeated with the large field in the left direction, followed by ever increasing field pulses to the right.

If the particle appeared to be multidomained, the procedure was as for a single-domain particle, except that in order to start each pulse to the right from approximately the same state of magnetization, the large field pulse applied each time to the left was replaced with a decaying oscillatory field. Such a field essentially demagnetizes the particle and leaves it in a nonmagnetic state.

Previous theoretical calculations⁷ indicate that a Fe_3O_4 or $\gamma\text{-Fe}_2\text{O}_3$ particle of prolate spheroidal shape, with a major to minor axis ratio of about 5, and about 1μ in length, will be single domained. On the other hand, a spherical particle of these materials would be multidomained if larger than $\approx 0.05\mu$ in diameter. We had available powders⁷ of magnetite or gamma ferric oxide, containing (i) acicular particles of length about 1 to 2μ and with major to minor axis ratio ranging from 8/1 to 6/1, (ii) cubical particles about 1μ in diameter, and (iii) irregularly shaped particles whose sizes varied up to about 60μ in diameter.

Experimental results, obtained with the torsion balance, on particles of the aforementioned powders, are reported in the next section.

RESULTS

The results on one of the irregular shaped particles of Fe_3O_4 , about 60μ in diameter (curve A), and on one of the cubical 1μ particles of Fe_3O_4 (curve B), are shown in Fig. 3. It is clear that the amount of remanent magnetization of each particle depended in a monotonic continuous fashion on the size of the applied field pulse. One can conclude that both these particles were multidomained, in accord with theoretical predictions.⁷ It is to be noted that the actual deflections observed for the two particles were about equal in magnitude. This resulted because (a) the gradient of the magnetic field, used for the deflection, was about twenty times greater, and (b) the diameter of the suspension fiber was about five times smaller, for the cubical particle than for the large angular one. Further, the maximum equivalent deflection for the cubical particle was about six times the maximum calculated¹ for a single-domain acicular particle, assumed to be a prolate spheroid of 1.0μ

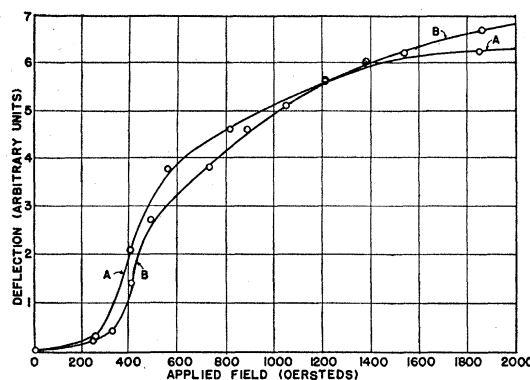


FIG. 3. The deflections of (A) a 60μ irregularly shaped particle of Fe_3O_4 , and (B) a cubical 1μ particle of Fe_3O_4 , as a function of a previously applied magnetic field pulse.

major axis length and 0.2μ minor axis length. This is reasonable, even though the volume of the cubical particle is about twenty-five times that of the acicular one, since the remanent magnetization of a multidomain particle is expected to be considerably less than the saturation magnetization. For the most favorable case, the remanent magnetization of a single-domain particle is equal to the saturation magnetization.

Next, measurements on one of the acicular particles of $\gamma\text{-Fe}_2\text{O}_3$ described earlier, are shown in Fig. 4. Since both theory⁷ and preliminary measurements indicated this to be a single-domain particle, the pulse procedure for a single-domain particle was used to obtain the illustrated data. It is obvious that the remanent magnetization remained constant until a field pulse of about 550 oersteds was applied. Such a field caused a small discontinuous change of the remanent magnetization. Further, a field pulse of about 800 oersteds caused another larger discontinuous change in the remanence. The remanence then remained constant with larger magnetic field pulses.

There are two possible explanations for the double "jump" in the remanence. One is that there were in reality two particles on the balance. Even with the highest magnifications possible with an optical microscope, it is difficult to be sure that only one particle has been isolated because of diffraction effects. The second possibility arises if an easy direction of magnetization for crystalline anisotropy is not along the easy direction for the shape anisotropy. This means that instead of there being only two directions of energy minima, and therefore only two directions along which the magnetization vector can lie, there may be four, or even more directions along which there are energy minima. However, we have plotted the magnetostatic and the anisotropy energies for various relative orientations for an acicular particle of Fe_3O_4 . In this case it seems that the shape anisotropy so predominates that it swamps the crystalline anisotropy. It therefore seems unlikely that there are more than two stable positions for the magnetization vector. We conclude

therefore, that the results of Fig. 4 are due to two particles, each of which is a single domain.

It will be observed that there is an asymmetry in Fig. 4. This is due to the fact that there are two torques acting on the particle during the deflection, one due to the gradient of the applied magnetic field, and the other due to $\mathbf{m} \times \mathbf{H}$. The torques add in one direction, and subtract in the other.¹ Finally, the magnitude of the observed deflection is in accord with that predicted by the calculations for an acicular particle¹ of this size.

Unfortunately, at this point in our experiments, we depleted the original supply of the quartz with which we had fabricated the suspension system of the torsion balance. This quartz had no detectable magnetic impurities. All further supplies we have been able to obtain up to the time of writing had some magnetic impurity. The result is that the deflection due to the impurity is usually several times that of one of the acicular particles, and consequently the discontinuous

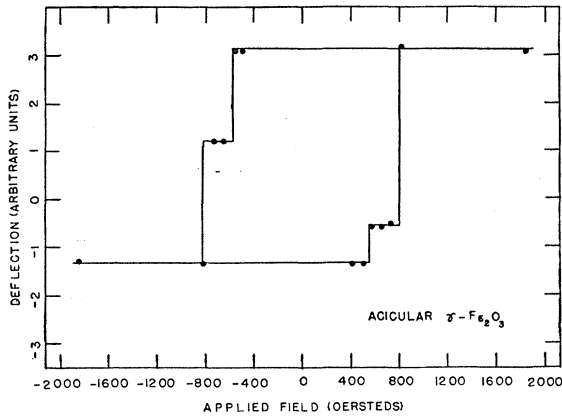


FIG. 4. The deflections of an acicular micron-sized $\gamma\text{-Fe}_2\text{O}_3$ particle, produced by applying an inhomogeneous magnetic field as a function of a previously applied magnetic field pulse.

jump in magnetization of the single-domain particle is not detectable.

However, we have fortunately succeeded in constructing two suspensions from the impure quartz in which the deflections due to the impurity and due to an acicular particle have been about the same magnitude. It is not clear whether these results have been achieved because of a smaller amount of impurity in the quartz of these particular suspension systems, or whether by chance the effect of one arm of the "T" has balanced out the effect of the other.

The results for one of these suspensions is shown in

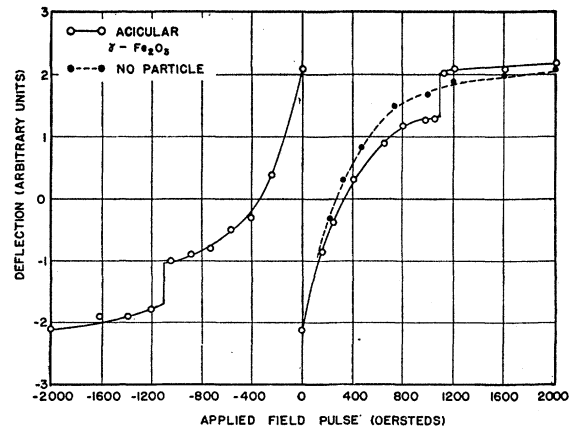


FIG. 5. The deflection of a quartz suspension system with and without an acicular $\gamma\text{-Fe}_2\text{O}_3$ particle.

Fig. 5. The full curve shows the deflection with the particle glued on the balance. A field pulse of 1100 oersteds causes a discontinuity in the remanence. Further, since the field pulse at which the discontinuity occurs should depend on the angle the major axis of the acicular particle makes with the magnetic field, the suspension system was rotated about 22° . It was then observed that the discontinuity in the remanence occurred at about 1000 oersteds. (These results have not been plotted in Fig. 5). Finally, the broken curve of Fig. 5 shows the deflection that resulted after the acicular particle had been clipped off the "T."

Similar results to those of Fig. 5 were obtained for another suspension.

Therefore, in spite of the impurity in the quartz, additional evidence has been obtained which strengthens the conclusion that the acicular particles are single-domain.

CONCLUSION

The existence of single-domain magnetic particles has been investigated in a very direct way. The results give strong evidence for the existence of single-domain particles. Further, the theoretical value of the critical size for single-domain behavior is roughly confirmed.

ACKNOWLEDGMENTS

We are greatly indebted to E. W. Davis, H. E. Hartig, W. G. Shepherd, H. H. Wade, and W. W. Wetzel for their support, interest, and assistance. The research was financed by funds from the Mines Experiment Station to the Department of Electrical Engineering.