Optical and Magnetic Properties of a Magnetite Suspension

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A suspension of magnetite powder in oil acts in a limited way as a light shutter. When a magnetic field is parallel to the light direction more light is transmitted than when the light is transverse. Microscopic examination shows that the particles form elongated groups. Each particle is probably a single magnetic domain, magnetized to saturation, and hence it attracts neighboring particles. The theory of the phenomenon, assuming the groups to be uniform and cylindrical, gives an equation for the amount of transmitted light in terms of magnetic field strength. This equation is subjected to experimental test and is not well verified. The magnetization curve of a dense suspension is determined experimentally and found to fit the Langevin curve fairly well. The group of particles, constituting the magnetic element, has an intensity of magnetization considerably less than the saturation value, although the individual particles are probably single, saturated domains.

IF rouge or Venetian red is strongly heated with a Bunsen flame the powder becomes more strongly magnetic. A little of this powder, shaken up in water or oil and placed in a glass cell, shows an increase in transmission of light if a magnetic field acts parallel to the light beam. Conversely there is a slight decrease of transmission for a transverse field. A microscopic examination of the suspension shows that the magnetic particles form small groups or chains. These groups when oriented by a magnetic field so as to lie parallel to the light beam afford less blocking area to the light than when randomly oriented.

THEORY

Suppose the suspension to consist of N similar cylinders contained in a cubical cell of unit volume. Let A be the area of the longitudinal median plane and E the area of the base of each cylinder. Let a magnetic field H and a light beam be parallel to the x axis. A cylinder whose axis makes an angle θ with the x axis will then cast on the yz plane a shadow of area $A\sin\theta+E\cos\theta$. Let dN be the number of cylinders per unit volume whose axes have orientations between θ and $\theta+d\theta$. Then the total area of the shadow cast on unit area of the yz plane will be

$$S=\int^{\pi}(AdN\sin\theta+EdN\cos\theta).$$

Here the suspension is assumed to be so dilute that superposition of shadows is negligible.

The value of dN will be given as in Langevin's theory of a paramagnetic gas by $dN=2\pi Ce^{a\cos\theta}\sin\theta d\theta$, where $C=Na/(4\pi\sinh a)$, $a=\mu H/kT$, $\mu=$magnetic moment of a cylinder, $k=$Boltzmann's constant, $T=$absolute temperature. The first term of the integral above is not easily evaluated when this value of dN is substituted and when a is large as in the present case. However, an approximately correct result can be obtained as follows.

It can be shown that

$$\int_0^{\pi}e^{a\cos\theta}\sin^3\theta d\theta=\sinh a(\coth a-1/a)4/a^2. \quad (1)$$

We now write

$$\int_0^{\pi}e^{a\cos\theta}\sin^3\theta d\theta=\langle\sin\theta\rangle_{\text{Av}}\int_0^{\pi}e^{a\cos\theta}\sin^2\theta d\theta, \quad (2)$$

where $\langle\sin\theta\rangle_{\text{Av}}$ represents an average value of $\sin\theta$. The actual average value of $\sin\theta$ is given by

$$(1/N)\int_0^{\pi}dN\sin\theta.$$

Substituting this value for $\langle\sin\theta\rangle_{\text{Av}}$ and solving for the unknown integral we get

$$\int_0^{\pi}e^{a\cos\theta}\sin^2\theta d\theta=(2/a)(2L/a)^{\frac{1}{2}}\sinh a, \quad (3)$$

where L is the Langevin function, $L=\coth a-1/a$. A rough graphical integration of the function for a specific value of a showed that Eq. (3) was not greatly in error.

The second term in the equation for S is easily integrated and the complete expression becomes

$$S=NA(2L/a)^{\frac{1}{2}}+NEL. \quad (4)$$

The value of a is presumably quite large. For a cylinder 2 microns in diameter and 3 microns long the value of a at 300°C is of the order 10^5H, assuming the saturation intensity of magnetization of magnetite to be 450. It is therefore possible to approximate and write $L=1-1/a$. In this case we have

$$S=NE-NE/a+NA(2/a)^{\frac{1}{2}}. \quad (5)$$

If l_0=amount of light transmitted by the unit cell before the particles are put in we have for the amount of light getting through the cell, $l=l_0(1-S)$. Thus

$$l/l_0=1-NE+NE/a-NA(2/a)^{\frac{1}{2}}. \quad (6)$$

Let $l=l_{||}$ when H is very large. In practice when H exceeded a few gauss the amount of light transmitted was not appreciably increased by further increase of H. Also let $l_\perp$ be the light transmitted when this saturation field acts at right angles to the light beam. We thus get $l_{||}/l_0=1-NE$ and $l_\perp/l_0=1-NA$. These equations allow the ratio E/A to be determined from measurements of light quantities. We may also eliminate the structure constants from Eq. (6), obtaining

$$(l_{||}-l)/(l_0-l_\perp)=(2/a)^{\frac{1}{2}}-(l_0-l_{||})/(l_0-l_\perp)a. \quad (7)$$

Unless H is small the second term on the right side may be neglected. From this equation it is possible to determine the magnetic moment of the magnetic particle by measuring light quantities in a known magnetic field.

If the Langevin theory applies to these particles in suspension we have $I/I_0=L$, where I is the intensity of magnetization of the suspension and I_0 is the saturation value of I. Using the approximation for L when a is large we have $1/a=1-I/I_0$. Substitution of this value in the above equation gives $I/I_0=1-r^2/2$, where $r=(l_{||}-l)/(l_0-l_\perp)$. It should thus be possible to determine the magnetization curve of the suspension by measuring light intensities in various fields.

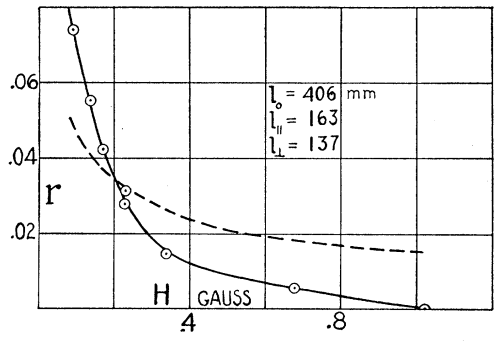

FIG. 1. Variation of $r=(l_{||}-l)/(l_0-l_\perp)$ with magnetic field. The dotted curve is given by the theory.

EXPERIMENTS

Preliminary experiments were made with jeweler's rouge, strongly heated with a Bunsen flame and then stirred up either in water or a mixture of light lubricating oil and kerosene. Because of the fact that this powder consists of a mixture of various oxides in unknown proportions the rouge was discarded and a fine powder made by grinding a small crystal of natural magnetite.

The powder so formed exhibited the Barkhausen effect when the particles were as much as 20μ in diameter, but when further grinding had reduced the diameter to about 10μ no Barkhausen effect could be detected. It is probable, therefore, that each particle of the finer powder consisted of a single magnetic domain, magnetized to saturation. (It is possible, of course, that grinding a particle containing several domains merely decreases the sizes of the domains without decreasing their number.[1]) Grinding was continued in an agate mill until the average particle diameter was less than 1μ.

When stirred up in light oil the particles stuck together and formed groups. Each particle seemed to behave in this respect like a tiny magnet. The suspension was allowed to stand for about half an hour so that the larger groups settled out, then the top portion was poured into a cubical glass cell. A beam of light made approximately parallel by a lens was sent through the liquid and allowed to fall on a photronic cell. Deflections of the galvanometer used with this cell were accurately proportional to light intensities. The earth's field was neutralized by

[1] J. Frenkel and J. Dorfman, Nature **126**, 274 (1930).

a pair of large Helmholtz coils, and a smaller pair was used to produce a magnetic field at the cell, either parallel or perpendicular to the light beam as desired. The value of $r=(l_{||}-l)/(l_0-l_\perp)$ was calculated for various fields, galvanometer deflections being taken as the measure of light intensity for the various cases.

Figure 1 shows the results obtained. The dotted curve is the graph of the equation $r=(2/a)^{\frac{1}{2}}$, where $a=8900H$. This equation, as shown above, should represent experimental results when a is not too small. There is very definite disagreement between theory and experiment.

The following causes of the discrepancy may be considered.

(1) Superposition of shadows may occur. The error caused by such overlapping of shadows of the different elements cannot be very large because experiments with suspensions of different densities gave essentially the same course for the curve. For one suspension the light transmitted by the cell was 73 percent of the incident light, for a second suspension, 35 percent. The curves for these two suspensions could be made to superpose within the limits of error of the experiment by multiplying r for one of the suspensions by a suitable constant factor. Thus we conclude that the average size of groups was different for the two specimens but the law of variation of light with H was the same.

(2) The orienting elements are certainly not all of the same size, hence the value of μ may vary for the different elements. This distribution in size of μ will not change the way in which r varies with H. Let dS_1 be the shadow cast by the small group dN_1 of the elements with moments comprised in the range μ_1 to $\mu_1+d\mu_1$. We thus get $dS_1=dN_1E_1+dN_1A_1(2/a)^{\frac{1}{2}}$, where the approximation is used for L when a is large. A similar expression is obtained for all the other groups into which the suspension may be divided. Summing up the shadows for all the groups gives the resulting shadow for the entire suspension. Since $a_1=\mu_1H/kT$ we have

$$S=\sum dS_1=\sum dN_1E_1+(2kT/H)^{\frac{1}{2}}\sum dN_1A_1/\mu_1^{\frac{1}{2}}.$$

Since $l=l_0(1-S)$, we get as before,

$$l/l_0=1-\sum E_1dN_1-(2kT/H)^{\frac{1}{2}}\sum dN_1A_1/\mu_1^{\frac{1}{2}}$$
$$=l_{||}/l_0-C/H^{\frac{1}{2}},$$

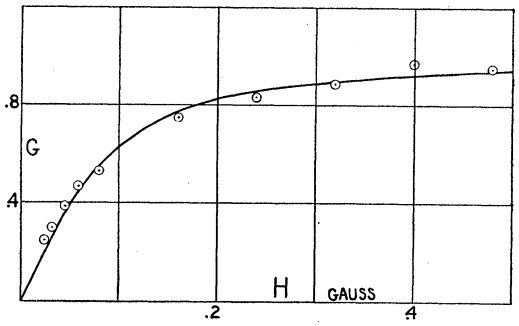

FIG. 2. Magnetization curve of magnetite suspension.

where C is a constant, independent of H. This last equation, therefore, gives $r=B/H^{\frac{1}{2}}$, where B is a constant. Thus it appears that r varies with H as indicated by Eq. (7) when a is large.

(3) The character of the suspension may change during the course of the experiment because of the settling out of the larger magnetic groups. To eliminate this error as far as possible the experiment was performed rapidly and the various magnetic fields were chosen in random order.

Since the theory does not agree with the experimental results we cannot use the equation $I/I_0=1-r^2/2$ for determining the magnetization curve of the suspension. To get this curve an apparatus similar to that used by Elmore[2] was set up. In this arrangement the presence of a test-tube of the fluid in a pick-up coil causes an inductive kick of a galvanometer connected to the coil when a magnetic field is applied. However, with a galvanometer of high sensitivity and a pick-up coil of 10,000 turns having cross-sectional area of 6.9 cm^2 the intensity of magnetization of the suspension was too weak to produce a measurable effect.

A more dense suspension of the magnetite powder was therefore made up and its magnetization curve determined. This suspension was so dense as to be absolutely opaque to light. Before each observation of the galvanometer deflection, produced by a given field, the tube of material was vigorously stirred. The various fields were applied with the same interval of elapsed time after the stirring, and random ordering of the various field strengths was used in getting the curve.

[2] W. C. Elmore, Phys. Rev. 54, 1092 (1938).

The points plotted on Fig. 2 represent the experimental results; the curve drawn is the graph of $G=G_0L$, where $G_0=1.03$, and the value of a in the Langevin function L is set equal to $25H$. Here G and G_0 represent galvanometer deflections, which are proportional to intensities of magnetization. It appears that the Langevin function represents the experimental results within the limits of experimental error. However the value of μ determined from these results is smaller than might be expected. Since $\mu/kT=25$ and $T=300°\text{K}$ we get $\mu=1.0\times10^{-12}$. The magnetic elements range in diameter from 1 to 10 microns. If the average is taken as 5 microns the intensity of magnetization of the element is approximately 0.015. For a large magnetite crystal the saturation intensity of magnetization[3] is about 450.

It is interesting to note that the value of μ for the less dense suspension used in the light experiment may be considerably larger. If we take $a=8900H$, the value appropriate to the dotted curve of Fig. 1, we get $\mu=3.6\times10^{-10}$. The magnetic elements were decidedly smaller in this suspension because the larger groups were allowed to settle out. Thus the intensity of magnetization of an element may here approach more nearly the value for the material in bulk.

Previous workers in this field have used colloidal solutions where the particles are much smaller than in the present case. Elmore,[2] using a magnetite colloid, found a discrepancy between the experimental curve and the Langevin curve, but he was able to explain the discrepancy by assuming a distribution in size of the particle moments. He also found a value of I_0 which was smaller than the value computed from the colloid concentration and intensity of magnetization of the magnetite in bulk. Elmore suggests as the most likely explanation of the latter discrepancy the presence of nonferromagnetic oxides of iron in his colloid, and states that it is fair to conclude that the small particles of his solution are not demagnetized in the macroscopic sense.

In the present experiments with larger particles we can still conclude that each particle is a single domain magnetized to saturation. The groups formed in the suspension constitute the magnetic element for which μ is determined. To explain the low value of μ in the dense suspension we assume each group to contain a sufficient number of particles, arranged in a sufficiently random manner, so that the average intensity of magnetization of the group falls considerably below that of the individual particles. The group retains a constant moment in the magnetic field because this field, though capable of orienting the group as a whole against the weak forces of viscosity, is unable to turn the separate particles out of the equilibrium positions which they assume in their own intense local fields.

There is certainly a rather wide distribution of μ values for the groups in this experiment. It is therefore not clear why the Langevin curve should be followed so much more accurately in the present experiment than in Elmore's experiment with colloids. As a matter of fact, close inspection of Fig. 2 gives some evidence of a slight departure in the direction indicated by Elmore's experiment. Such a departure, however, is almost masked by experimental error.

As regards the transmission of light through these suspensions we must conclude that the theory as given above contains approximations or simplifications which are too crude to allow good agreement with experimental results. A more accurate theory, which allows for the presence of spherical groups, or which considers diffraction effects, would give an equation somewhat different from Eq. (7). However, such an equation still fails to give the correct law of variation of light with magnetic field.

[3] P. Weiss and R. Forrer, Ann. de physique **12**, 330 (1929).