LA-UR 91 - 3851

LA-UR--91-3851

DE92 005018

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TITLE PROBING FINE MAGNETIC PARTICLES WITH NEUTRON SCATTERING

AUTHOR(S) R. Pynn, LANSCE

SUBMITTED TO International Workshop On: Studies Of Magnetic Properties Of Fine
Particles And Their Relevance To Materials Science
November 4-8, 1991 in Rome Italy

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Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Probing Fine Magnetic Particles with Neutron Scattering

R. Pynn

Manuel Lujan, Jr. Neutron Scattering Center, Los Alamos National Laboratory,
P.O. Box 1663, MS H805, Los Alamos, New Mexico 87545, USA

Abstract

Because thermal neutrons are scattered both by nuclei and by unpaired electrons, they provide an ideal probe for studying the atomic and magnetic structures of fine-grained magnetic materials, including nanocrystalline solids, thin epitaxial layers, and colloidal suspensions of magnetic particles, known as ferrofluids. Diffraction, surface reflection, and small angle neutron scattering (SANS) are the techniques used. With the exception of surface reflection, these methods are described in this article. The combination of SANS with refractive-index matching and neutron polarisation analysis is particularly powerful because it allows the magnetic and atomic structures to be determined independently. This technique has been used to study both dilute and concentrated ferrofluid suspensions of relatively monodisperse cobalt particles, subjected to a series of applied magnetic fields. The size of the cobalt particle core and the surrounding surfactant layer were determined. The measured interparticle structure factor agrees well with a recent theory that allows correlations in binary mixtures of magnetic particles to be calculated in the case of complete magnetic alignment. When one of the species in such a binary mixture is a nonmagnetic, cylindrical macromolecule, application of a magnetic field leads to some degree of alignment of the nonmagnetic species. This result has been demonstrated with tobacco mosaic virus suspended in a water-based ferrofluid.

1. INTRODUCTION TO NEUTRON SCATTERING

In 1936, Fermi [1] published a theoretical description of the scattering of slow neutrons by nuclei and introduced the concept of a scattering length, b , that completely describes the neutron-nuclear interaction. In Fermi's picture, the strong, spatially dependent interaction between the nucleus and a neutron is replaced by a delta-function pseudopotential, $U(r)$, of magnitude b that can be used in a perturbation calculation of the scattering. According to a later analysis performed in 1954 by Van Hove [2], the probability of a neutron being scattered coherently and elastically by an assembly of nuclei located at positions r_j is given by

$$I_{\text{coh}}(\mathbf{Q}) = \sum_{j,k} b_j b_k \langle e^{i\mathbf{Q}\cdot\mathbf{r}_j} e^{-i\mathbf{Q}\cdot\mathbf{r}_k} \rangle \quad (1)$$

The sum is over pairs of nuclei j and k , with the nucleus labelled j , of coherent [3] scattering length b_j , at position r_j . The angular brackets denote the usual thermodynamic average or, equivalently, an average over all possible starting times for observation of the system. The quantity $\hbar Q/2\pi$ is the momentum transferred to the sample during the neutron scattering process. It is given, in terms of the wavevectors, k_i and k_f , of the incident and scattered neutrons by the conservation law,

$$Q = k_i - k_f \quad (2)$$

The nuclear scattering length, b , may be either positive or negative (negative values correspond to a phase shift of π on scattering) and has a magnitude that appears to vary, apparently at random, from isotope to isotope. Of particular interest are the values for hydrogen and deuterium: -3.741 and $+6.674$ Fermis ($1 \text{ Fermi} = 10^{-15} \text{ metres}$), respectively. Because deuterium can often be substituted for hydrogen in organic molecules, the scattering power of the molecules for neutrons may be altered by this means, allowing particular components of a molecule to be highlighted for neutron scattering.

The year after Fermi published his pseudopotential, Schwinger [4], analysed theoretically the interaction of the neutron's magnetic moment with a ferromagnetic material and correctly predicted the angular dependence of the scattering. In this case, the neutron is scattered by the distribution of magnetic induction, B , within a material, and thus probes the density of unpaired electron spins. The scattering length for magnetic scattering of neutrons, p , is a calculable quantity:

$$p = \gamma r_e |M| f(Q)/2 \quad (3)$$

where r_e ($=2.818 \times 10^{-15} \text{ m}$) is the classical electron radius, M is the total electronic magnetic moment in Bohr magnetons, $f(Q)$ is the electronic form factor evaluated at the wavevector Q , and γ ($=+1.913042$ nuclear magnetons) is the *magnitude* of the magnetic moment of the neutron. Because the magnetic interaction between neutrons and matter is dipolar in origin, only the component of the sample magnetisation, M , perpendicular to the wavevector transfer, Q , is effective in scattering neutrons. To calculate the total, elastic, magnetic scattering from a macroscopic sample, one simply replaces b_j in eq. (1) with the appropriate value of p_j for the j^{th} atomic site. These values of p_j are obtained by multiplying p [cf. eq. (3)] by a unit vector, $m_{\perp}$, that is colinear with the projection of M on the plane perpendicular to Q .

An evaluation of eq. (3) shows that nuclear and magnetic scattering of neutrons are of comparable magnitude: p takes a value of about 2.7 Fermis for an electronic moment of one Bohr magneton compared to the value of -3.741 Fermis quoted above for the coherent scattering length of a hydrogen nucleus. For this reason, neutron scattering provides an ideal tool for measuring both the atomic and magnetic structures of matter.

In general, neutrons are scattered at the same value of Q by variations of atomic and magnetic densities in a sample, and it can be difficult to distinguish one source of scattering from the other unless a further trick, known as neutron polarisation analysis, is used. First, a beam of neutrons in which the neutron magnetic moments are all aligned in one direction is prepared [5]. A magnetic field of a few Oersteds applied to the neutron beam maintains the direction of the neutron moments and prevents beam depolarisation. This field is known as a guide field. Depending on the nature of the scattering process, the neutron spin (which is antiparallel to its magnetic moment) either retains its orientation with respect to a magnetic field applied to the scattering sample or is reversed. Scattering events

for which the spin is reversed are referred to as *spin-flip* (sf) processes, whereas those for which the spin direction remains unchanged are *non-spin-flip* (nsf). A number of simple rules govern the various possible neutron scattering processes:

- (i) nuclear coherent scattering [cf. eq. (1)] is always *non-spin-flip*;
- (ii) the component of $\mathbf{m}_\perp$ that is parallel to the magnetic field applied to the scattering sample is associated with *non-spin-flip* scattering; and
- (iii) components of $\mathbf{m}_\perp$ that are perpendicular to the magnetic field applied to the scattering sample are associated with *spin-flip* scattering.

More formally, we append to the Fermi pseudopotential, U , one of the combinations: $(++)$, $(--)$, $(+-)$, and $(-+)$, the first two of which represent *non-spin-flip* scattering and the last two, *spin flip* scattering. The combination $(+-)$, for example, means that a neutron is scattered from a state $(+)$, in which its spin is parallel to the applied magnetic field, $\mathbf{H}$, to a state $(-)$, where the neutron spin is antiparallel to $\mathbf{H}$. Moon, Riste, and Koeler [6] showed that, in the absence of incoherent scattering,

$$U^{++} = b - p m_\perp^z \quad (4a)$$

$$U^{--} = b + p m_\perp^z \quad (4b)$$

$$U^{+-} = -p(m_\perp^x + i m_\perp^y) \quad (4c)$$

$$U^{-+} = -p(m_\perp^x - i m_\perp^y) \quad (4d)$$

where the z Cartesian axis is parallel to $\mathbf{H}$. To determine the scattered intensity measured in a particular polarisation analysis experiment, the scattering length in eq. (1) is replaced by the appropriate value of U from eq. (4). When this is done, eqs. (4a) and (4b) imply rules (i) and (ii) above, while eqs. (4c) and (4d) imply rule (iii).

By measuring *spin-flip* and *non-spin-flip* scattering for various orientations of a magnetic field applied to the scattering sample, magnetic and nuclear contributions to neutron scattering can often be separated. For example, if the guide field is parallel to $\mathbf{Q}$, *non-spin-flip* scattering can only be of nuclear origin, according to the rules given above. We shall discuss an example of the use of polarisation analysis in connection with experiments to determine the structure of ferrofluids.

The subject of this workshop is fine magnetic particles. To study the overall structure, as opposed to the atomic arrangement, of objects in the $10\text{\AA}$ to $1000\text{\AA}$ range with neutron scattering requires special instrumentation and a technique known as small angle neutron scattering (SANS) [5]. That the length scales probed by neutron scattering depends on the angle through which neutrons are scattered can be understood from Figure 1. For a given neutron wavelength, λ , the size of objects probed increases as the scattering angle decreases. Because

$$|\mathbf{Q}| = |\mathbf{k}_i - \mathbf{k}_f| = \frac{4\pi}{\lambda} \sin \theta \quad (5)$$

where 2θ is the scattering angle (angle between $\mathbf{k}_i$ and $\mathbf{k}_f$), Q decreases as the scattering angle decreases and the neutron wavelength increases.

Figure 1. One way to view neutron scattering is to imagine that the incident and scattered neutron waves form a secondary "probe wave" (here seen as a Moiré pattern) whose wavelength determines the length scale examined in the sample. The two pictures clearly show that a reduction of the scattering angle corresponds to an increase in the size of objects probed.

For a system composed of independent similar particles, such as a colloidal dispersion, each sum in eq. (1) may be separated into a sum over atoms within a particle and a sum over particles. For sufficiently small values of Q , the phase factors in eq. (1) vary slowly from atom to atom within a particle, so the scattering probability at small Q can be written as

$$I^{ss'}(Q) = S(Q) |F^{ss'}(Q)|^2 \quad (6a)$$

$$\text{where } F^{ss'}(Q) = \int_{\text{particle}} U^{ss'} e^{iQ \cdot r} d^3r \quad (6b)$$

where ss' denotes one of the neutron spin combinations in eq. (4). In eqs. (6), $F^{ss'}(Q)$ is the particle form factor and $S(Q)$ the structure factor that measures correlations between the particles of the system [7].

2. APPLICATIONS OF NEUTRON SCATTERING

2.1. Nanocrystalline Chromium

Equation (1) is the basis for determining atomic and magnetic structures by neutron diffraction. When all of the atoms (or magnetic moments) in a material are arranged on a regular lattice, the scattered neutron intensity vanishes except when Q is equal to a

reciprocal lattice vector of the structure. All of the neutrons are scattered in well-defined directions to give a series of Bragg reflections whose intensities provide information about the atomic (or magnetic) ordering within a unit cell. If the scattering sample is large, each measured Bragg peak is a Dirac delta function convolved with the resolution of the neutron spectrometer used to measure it. In practice, for measurements of diffraction from a powder sample, the resolution width, ΔQ_{res} , may achieve values of less than $10^{-3}Q$ with modern neutron spectrometers. In fine grained, monodisperse materials, the sum in Eqn. (1) is cut off at the edge of a particle so that Bragg peaks have a natural width, δQ , that is equal to $2\pi/d$, where d is the particle size. In practice, most samples contain particles with a range of sizes so that the widths of Bragg peaks measure an average particle diameter. Nevertheless, Bragg peaks whose natural widths do not vary with Q are a clear signature of diffraction by small particles. Lattice strain, for example, introduces a broadening that is proportional to the value of Q . Experiments to examine the magnetic structure of nanocrystalline chromium have been undertaken by Fitzsimmons et al. [8], who intend to use the relative intensities of nuclear and magnetic Bragg peaks to obtain the magnetisation of chromium ions and the widths of Bragg peaks to deduce the fraction of each chromium crystallite that scatters magnetically. Preliminary data indicate that the spin-density wavevector in nanocrystalline chromium is displaced from that in bulk material.

2.2. Dilute Ferrofluids

Colloidal dispersions of magnetic particles, known as ferrofluids and stabilized by the addition of a surfactant, form a technologically important class of magnetic fluids with novel structural features [9]. In a sufficiently dilute ferrofluid, there is no correlation between individual colloidal particles, so the structure factor, $S(Q)$ in eq. (6), is unity for all Q . Thus, a SANS experiment measures some combination of magnetic and nuclear form factors for the particles and can be used to deduce the structure of individual particles. From eq. (5), it is easy to show that the nsf scattering for the cases when Q is either parallel or perpendicular to H is given by

$$I(Q\parallel H) = F_N^2(Q) = \left| \int_{\text{particle}} b(r) e^{iQ \cdot r} d^3r \right|^2 \quad (7a)$$

$$I(Q\perp H) = [F_N(Q) \pm F_M(Q)]^2 \quad (7b)$$

$$\text{where } F_M(Q) = \int_{\text{particle}} p(r) e^{iQ \cdot r} d^3r \quad (7c)$$

Polarisation analysis thus allows the magnetic and atomic structures of ferrofluid particles to be probed independently. Furthermore, because the component of ferrofluid magnetisation perpendicular to H give rise to spin flip scattering, neutron polarisation analysis also provides a measure of the degree of magnetic alignment achieved in the ferrofluid.

Equation (7) was derived on the assumption that all colloidal particles are the same shape and size. This is not always the case. The practical implications of such polydispersity can be quite severe, as Nunes has demonstrated [10]. In the experiments reported here [11], great pains were taken to use relatively monodisperse samples. Electron micrographs indicated that the cobalt particles used were approximately spherical, with an average radius of 28 ± 3 Å. The distribution of core radii about the mean value has

a half-width of 6 Å, and numerical calculations indicate that this polydispersity does not affect significantly the conclusions drawn from our neutron scattering measurements.

An evaluation of the atomic and magnetic form factors, $F_N(Q)$ and $F_M(Q)$, requires a model of the ferrofluid particle. For the nuclear scattering, the simplest model is a uniform, spherical core surrounded by a uniform shell of surfactant. The magnetic structure is entirely due to the spherical metallic core. Nuclear and magnetic scattering length densities, $b(r)$ and $p(r)$, for the cobalt core, surfactant shell, and solvent are shown in Figure 2. The scattering length density for the surfactant has been calculated under the assumption of a uniform packing of surfactant molecules. The solvent used in the experiments was chosen to have a lower scattering length density than that of the surfactant: the opposite choice leads to significant structure in $F_N(Q)$ which makes it difficult to determine $S(Q)$ from data obtained with a concentrated ferrofluid.

Figure 2. Neutron scattering length densities as a function of radius for a cobalt ferrofluid particle. The radii are deduced from a best fit to data described in the text.

Figure 3: Measured values of the nsf scattering as a function of Q for a dilute (0.014 volume fraction) colloidal suspension of cobalt particles in dodecane, stabilised by Monoxol-OT and Imidrol-OC surfactants in a 3:1 ratio. The applied field was 0.5 Tesla. The line is calculated from fits to eqs. (7). (a) $Q \perp H$, (b) $Q \parallel H$.

The measured nsf scattering that represents the two cases described by eq. (7) is shown in Figure 3, for a cobalt ferrofluid of volume fraction 0.014 in an applied field of 0.3 Tesla. Since the volume fractions of cobalt and surfactant used in the preparation of the ferrofluid are known, there is a known relation between R_{core} and R_{tot} (cf. Figure 2 for definitions) if all the surfactant is on the cobalt particles. Because the data for $Q \parallel H$ only involve

nuclear scattering, they can be fit by two parameters: a scaling factor and R_{tot} . The result of this fit is shown by the lower solid curve in Figure 3.

One may ask whether a different model could provide an equally good description of the $Q \parallel H$ data. For example it is conceivable that all of the surfactant head groups are localised on the core. If this were the case, surfactant tails and the solvent would have similar scattering length densities. The value of R_{tot} found for such a model is about 50 Å. However, this value is completely at odds with the position of the first peak in data obtained with a concentrated dispersion (cf. Figure 4). A similar inconsistency is observed if one assumes that less than 80% of the total surfactant is associated with the cobalt particles. Thus, in spite of its apparently simple form, the form factor can provide quite detailed information about the ferrofluid particles.

Once the atomic structure of the ferrofluid particles has been determined, the only unknown quantity to be fit to the $Q \perp H$ data is R_{mag} . This turns out to be about 2.5 Å less than R_{core} , apparently indicating a non-magnetised layer of cobalt at the surface of the metallic core. However, it turns out that one can, equally well, fit the data by assuming that the magnetisation of the core is less than that of bulk cobalt [12], an explanation that is probably more likely in view of other evidence [13].

2.3. Concentrated Ferrofluids

A model for the structure factor of a binary mixture of fully aligned magnetic particles has been developed by Hayter and Pynn [14]. This calculation, which involves no adjustable parameters, can also be applied to a single component, dense ferrofluid. Direct comparison with experiment can thus be made, once the scale factor has been determined by matching theory and experiment at the peak value of $I_{\perp}(Q)$ for the largest applied magnetic field.

Figure 4a shows the nsf scattering obtained with a cobalt ferrofluid of volume fraction 0.27 in a field of 0.5 Tesla. Two theoretical curves are included, corresponding to the inclusion (full curve) or exclusion of short-range, attractive central forces modeled by a Yukawa potential of depth 0.7 k_BT and range 0.4 R_{tot} . Such particle "stickiness" has little effect on agreement between experiment and theory, which is good in either case.

Figure 4: Measured values for the nsf scattering (filled circles) and fluctuation scattering (open circles) for a ferrofluid at 0.27 total volume fraction in an applied magnetic field of (a) $H = 0.5$ Tesla; (b) $H = 0.2$ Tesla. Calculated lines are discussed in the text.

(i) $Q \perp H$, (ii) $Q \parallel H$.

The open symbols in Figure 4a are obtained from the measured spin-flip scattering as

$$I_{\text{fluc}}(Q) = 3 [I_{\text{sf}}(Q \parallel H) - I_{\text{sf}}(Q \perp H)] \quad (8)$$

Nuclear-spin incoherent scattering is the same in both field geometries and so cancels from this expression, which represents scattering from fluctuations of magnetisation that are perpendicular to H . I_{fluc} has the same Q dependence as $F_M(Q)$ at all applied fields up to 0.5 Tesla, indicating that magnetic fluctuations are substantially paramagnetic. For such a system, a simple Langevin treatment yields:

$$I_{\text{fluc}}(Q) = 3 F_M^2(Q) L(m)/m \quad (9)$$

where $m = \mu H/kT$, with μ as the dipole magnetic moment of the cobalt particles, and the Langevin function is:

$$L(m) = \coth m - 1/m \quad (10)$$

As Figures 4a and 4b demonstrate, eq. (9) provides a good description of the fluctuation scattering for applied fields of 0.5 and 0.2 Tesla. Provided $S(Q)$ is isotropic (which is almost the case for the cobalt ferrofluid studied here), one can use the Langevin formalism to deduce an expression for the nsf scattering when a ferrofluid is not magnetically saturated:

$$I^{++}(Q \perp H) = [F_N(Q) + L(m) F_M(Q)]^2 S(Q) + F_{\text{fluc}}^2(Q) [1 - 2L(m)/m - L^2(m)] \quad (11)$$

At the highest field ($H = 0.5$ T), there is little difference between the results of eqs. (6a) and (11), justifying *a posteriori* the use of eq. (6a) even in the presence of measurable fluctuation scattering. For intermediate fields, eq. (11) provides a reasonable description of the nsf scattering (cf. Figure 4b).

2.4. Binary Mixtures of Magnetic and Non-Magnetic Colloids

In a fascinating series of experiments, Skjeltorp [15] has demonstrated a wide range of structural possibilities that exist when magnetic and nonmagnetic particles coexist in the same colloidal dispersion. In the cases described by Skjeltorp, ordering of the nonmagnetic species in an applied magnetic field arises because the nonmagnetic particles behave as "magnetic holes" in a magnetic continuum. The moment of hole is equal to the total moment of ferrofluid displaced by the nonmagnetic particle. This way of thinking about the physics of a binary mixture works because the nonmagnetic particles used by Skjeltorp are large, with diameters of a micron or more. John Hayter and I asked ourselves the question: "What might happen if the magnetic and nonmagnetic species were of roughly equal size?" We have addressed this question both theoretically and experimentally [14,16]. For the remainder of this paper, I will describe the experimental results we obtained by neutron scattering [16] in a collaboration that spanned four countries!

A series of experiments were carried out with rodlike macromolecules, with aspect ratios of order 10:1, mixed with a fully deuterated water-based ferrofluid that was chosen to avoid damage to the macromolecules. Deuteration ensured that the ferrofluid particles

were almost contrast matched with the solvent (i.e., they had similar scattering length densities) so that neutron scattering resulted principally from the nonmagnetic macromolecules. The macromolecules used were tobacco mosaic virus (TMV) and tobacco rattle virus (TRV). TMV is a hollow cylindrical assembly of length 3000 Å, external diameter 180 Å, and internal diameter 40 Å. SANS experiments were performed on TMV dispersed in ferrofluid at 0.06 volume fraction. In zero magnetic field, the neutron scattering pattern is isotropic and displays a peak at 1.73 Å^{-1} that is characteristic of correlations between TMV particles under the solution conditions used. In a modest applied field, the scattering becomes anisotropic, with a peak appearing only for Q perpendicular to H , as shown in Figure 5a. The viruses have apparently been aligned within the ferrofluid by the application of a magnetic field. This alignment cannot be due to the "magnetic hole" effect described above, because each virus displaces, on average, less than one ferrofluid particle. The alignment is believed to result from cross correlations between magnetic and non-magnetic species.

Figure 5. Blank-subtracted SANS data for TMV dispersed at 0.06 volume fraction in a water-based ferrofluid. The vertical and horizontal dimensions of each plot correspond to $2Q_{\text{max}}$, where (a) $Q_{\text{max}} = 0.02 \text{ Å}^{-1}$ and (b) $Q_{\text{max}} = 0.355 \text{ Å}^{-1}$. White corresponds to minimum and black to maximum scattered intensity.

At larger values of Q , peaks in the diffraction pattern reflect internal structure of the TMV particles themselves. This is shown in Figure 5b, where the higher- Q peaks now appear for Q parallel to H . These peaks correspond to a helical protein coating of the TMV molecules that has a pitch of 23 Å. From the angular width of these diffraction peaks, we deduce that the orientation distribution of the TMV particles has a half-width of about 19° , corresponding to a nematic order parameter of about 0.8. More detailed measurements of this system have been made recently by Sosnick et al. [17].

2.5. Conclusion

Because of its sensitivity to both magnetism and atomic density, the neutron is an ideal structural probe for magnetic materials over length scales that vary between 1 Å and 1000 Å.

2.6. Acknowledgements

Most of the work I have done on ferrofluids has been done with John Hayter in a collaboration that I have enjoyed over many years. This work was partially supported by the US Department of Energy under contract W-7405-ENG-36 with the University of California.

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