SANS contrast variation method applied in experiments on ferrofluids at MURN instrument of IBR-2 reactor

Maria Balasoiu\textsuperscript{1,2}, Alexander Kuklin\textsuperscript{2}
\textsuperscript{1}Horia Hulubei National Institute of Physics and Nuclear Engineering, Bucharest, Romania
\textsuperscript{2}Joint Institute of Nuclear Research, Dubna, Russia

E-mail: balasoiumaria@yahoo.com; balas@nf.jinr.ru

Abstract. Separate determination of the nuclear and magnetic contributions to the scattering intensity by means of a contrast variation method applied in a small angle neutron scattering experiment of nonpolarized neutrons in ferrofluids in early 90’s at the MURN instrument is reviewed. The nuclear scattering contribution gives the features of the colloidal particle dimensions, surfactant shell structure and the solvent degree penetration to the macromolecular layer. The magnetic scattering part is compatible to the models where is supposed that the particle surface has a nonmagnetic layer. Details on experimental “Grabcev method” in obtaining separate nuclear and magnetic contributions to the small angle neutron scattering intensity of unpolarized neutrons are emphasized for the case of a high quality ultrastabile benzene-based ferrofluid with magnetite nanoparticles.

1. Introduction

In the last more than four decades, studies of ferrofluids have attracted an increased interest (e.g. references [1-46]) due to their intrinsic physical properties of these materials as well as to their remarkable technological importance. Among some other experimental techniques (magnetization and hydrodynamic measurements, electron microscopy (EM), high resolution transmission electron microscopy (HRTEM), photon correlation spectroscopy, light scattering, X-ray small angle scattering, Mossbauer spectroscopy, inelastic scattering, neutron depolarization, muon spectroscopy, synchrotron radiation) the small angle neutron scattering (SANS) has been efficiently used for the determination of particle structure (see e.g. references [10-19]), structural and magnetic correlations (see e.g. references [10-35]), aggregation (see e.g. references [33-40]), dynamics (see e.g. references [20,41-43]), particle size distribution and polydispersity effects (see e.g. references [17,33-40,44,45]) in ferrofluids, of new stabilization compounds and methods [9,32,35, 46].

In the present paper, SANS experiments with the use of contrast variation method (for general information see references [47-49], studying the structure of magnetic colloidal particles, performed in 90’s on the small angle scattering facility MURN [50,51] (named YuMO lately [51-54]) installed at the IBR-2 high pulsed reactor of the Joint Institute of Nuclear Research, Dubna, Russia, are reviewed.

Resulting from these experiments, it was established that within the Guinier [55] scattering range where the magnetic form factor, \(f\), of the magnetite is approximately 1 with an error less than 1\% [56], using the contrast variation method - when nuclear contrast changes the sign inside the D/H...
concentration variation range, the contribution of nuclear and magnetic components of the experimental intensity can be obtained separately even in the case of the unpolarized neutrons [14-18]. This method now is known as the “Grabcev method” upon the name of the coordinator of the experiments.

2. Experimental

The magnetite ferrofluids were prepared [57] in D- and H-benzene with oleic acid surfactant. The size distribution obtained by EM analyzes showed log-normal form with a standard deviation of ln $R$ 0.3 and the most probable radius $R_0 = 5.8$ nm. By dilution with measured quantities of D-benzene and/or H-benzene volume fraction $f = 0.6\%$ was obtained. The D-benzene volume fraction in the carrier $x$ was produced to take six equidistant values in the range of 0.0 to 1.0.

The SANS experiments consisted in measuring, for each value of $x$, $I_f(Q)$, the intensity as a function of $Q$, the momentum transfer, $Q = (4\pi / \lambda) \sin(\theta / 2)$, where $\lambda$ is the wavelength of the neutron while $\theta$ is the scattering angle. Runs were also performed on all the compositions of the carrier liquids, free of particles, measuring in such way $I_c(Q)$. The determination from the experimental data of $d\Sigma_f(Q)/dQ$ and respectively $d\Sigma_c(Q)/dQ$, the differential elastic coherent scattering cross-sections per unit solid angle and unit sample volume, of the ferrofluids (subscript $f$) and, respectively, of the carrier liquid (subscript $c$), was performed according to the procedure described in [51]. The difference,

$$\frac{d\Sigma}{dQ}(Q) = \frac{d\Sigma_f}{dQ}(Q) - \frac{d\Sigma_c}{dQ}(Q),$$

represents the excess scattering power of colloidal particles.

3. Results and discussions

Neglecting interparticle correlations and size polydispersity, on one hand, and presuming approximately spherical particles, on the other, for values of $Q$ small enough to ensure the Guinier’s law to be valid and magnetite magnetic form factor $f \sim 1$, the differential scattering cross-section for unpolarized neutrons may be written as

$$\frac{d\Sigma}{dQ}(Q) = \frac{d\Sigma_n}{dQ}(Q) + \frac{d\Sigma_m}{dQ}(Q)$$

(1)

or, approximately,

$$\frac{d\Sigma}{dQ}(Q) \approx \frac{d\Sigma_n}{dQ}(0)\exp(-R_{gn}^2Q^2/3) + \frac{d\Sigma_m}{dQ}(0)\exp(-R_{gm}^2Q^2/3)$$

(2)

The first term in the right side of the above equations represents the nuclear component of the cross-section while the second, the magnetic one. The quantities $R_{gn}$ and $R_{gm}$ are corresponding gyration radii while

$$\frac{d\Sigma_n}{dQ}(0) = N_p\rho^{\ast 2}V_d^2$$

(3)

$$\frac{d\Sigma_m}{dQ}(0) = (2/3)N_p\rho^{\ast 2}_mV_m^2$$

(4)
and, $N_p = 3 \cdot 10^{15}$ particles/cm$^2$ is the particle concentration; $V_d$ - represents the volume of the magnetite particle with the adsorbed surfactant macromolecules on its surface; $V_m$ - represents the magnetic volume; it is considered having a constant magnetization value.

$\rho^*$ - represents the contrast [47-49]; to calculate its expression, following conditions are considered:

- The colloidal particle is central-symmetrical and composed from a magnetite core surrounded by a surfactant shell;
- The surfactant shell has a nonuniform structure; partially, it can be penetrated by the carrier and empty spaces;
- The scattering length densities inside the volumes occupied by magnetite particles, surfactant and carrier, are uniform.

and,

$$\rho^* = (1/V_d) \int [\rho_n(\vec{r}) - \rho_c] d\vec{r} =$$

$$= (4\pi/V_d) \int_0^{R_c} (\rho_{mm} - \rho_c) r^2 dr + \phi \int_{R_0}^{R_c} (\rho_r - \rho_c) r^2 dr + \gamma \int_{R_0}^{R_c} (\rho_{mm} - \rho_c) r^2 dr +$$

$$+ (1 - \phi - \gamma) \int_{R_0}^{R_c} (0 - \rho_c) r^2 dr = 1/V_d \{ (\rho_{mm} - \rho_c) V_0 + [\phi \rho_s - (1 - \gamma) \rho_c] V_s \}$$ (5)

$\rho_c$ is the scattering length density of the carrier:

$$\rho_c = x \rho_{C_6D_6} + (1 - x) \rho_{C_8H_8}$$

where $\rho_{C_6D_6}$ and $\rho_{C_8H_8}$ are the scattering length densities of deuterated and, respectively, hydrogenated benzene; $\rho_{mm}$ is the magnetic scattering length density of the magnetite core; $V_s$ is the total volume of a colloidal particle, including the surfactant layer; $V_m$ is the magnetic active volume, i.e. the volume with a nonvanishing magnetization density; the numerical ratio $2/3$ in equation (4) is the polarization factor for unpolarized neutrons.

In Figure 1, the radial distribution of $\rho_n$ (density of nuclear coherent scattering length), inside as well as outside a colloidal particle, is shown. The notations $\rho_{mm}$ and $\rho_c$ are the nuclear scattering length densities of magnetite and, respectively, oleic acid surfactant. The ultimate values of carrier scattering length density are shown to illustrate the basic concept of the contrast variation method. The radial distribution of $\rho_{mm}$, the magnetic coherent scattering length density for the Guinier region where the magnetic form factor $f \sim 1$ with an error less than 1%, is plotted in Figure 2. Here $R_m$, the magnetic radius of the magnetite core may be smaller than $R_0$. At the same time, there has been assumed that for $r < R_m$ the magnetization density acquires the same value as in the bulk state. For the calculation of $\rho_{mm}$, a mass density $d_m = 5.2$ g/cm$^3$ and a magnetic moment $\mu = 4.2$ Bohr magnetons/molecule [58] are considered for $Fe_3O_4$. 


Figure 1. Method of contrast variation: $\rho_n$ - is the radial distribution of the density of nuclear scattering of the particle; $\rho_s$ - is the radial distribution of the nuclear scattering density of the surfactant; $\rho_{C_6D_6}$ and $\rho_{C_6H_6}$ - are the radial distributions of the nuclear scattering density of the carrier - deuterate benzene, respectively of hydrogenate benzene.

Figure 2. Radial distribution of the magnetic scattering density of the particle - $\rho_m$ for the momentum transfer $Q = 0$, where the magnetic form factor has a maximal value.

The numerical values of scattering length densities (Figure 1) are:

$\rho_{mn} = 6.977 \cdot 10^{10} \text{ cm}^{-2}$; $\rho_s = 0.077 \cdot 10^{10} \text{ cm}^{-2}$; $\rho_{C_6H_6} = 1.182 \cdot 10^{10} \text{ cm}^{-2}$; $\rho_{C_6D_6} = 5.437 \cdot 10^{10} \text{ cm}^{-2}$ and $\rho_{mn} = 3.068 \cdot 10^{10} \text{ cm}^{-2}$.

In Figure 3, the $d\Sigma(0)/d\Omega$ obtained by extrapolation of experimental data, is plotted against $x$. A minimum appears here, in the vicinity of $x = 0.6$, one of the values used in the experiment for $C_6D_6$ volume fraction in the carrier. An inspection of equations (1)-(4) shows that only the first term, namely that corresponding to nuclear scattering, depends of $x$. One may conclude hence that at $x = x_0$, the particle contrast, $\rho^*$ as a linear function of $x$, changes the sign.
Figure 3. Dependence of $d\Sigma(0)/d\Omega$ on $x$, the D-benzene value fraction in the carrier. There appears a minimum at $x = 0.6$. The dashed line is just an approximate guide for the eye.

However, although theoretically it is not possible to equal rigorously to zero the contrast value of an inhomogeneous particle embodied in a homogenous medium, the basic hypothesis of the following model data treatment is that at $x = x_0$ the nuclear contrast, $\rho^*$, is negligibly small as compared with the value of the magnetic scattering length density, $\rho_{mm}$, so that at this point the residual contribution of nuclear scattering cross section may be disregarded, or in other words

$$\frac{d\Sigma}{d\Omega}(0) \bigg|_{x=x_0} = \frac{d\Sigma_m}{d\Omega}(0)$$  \hspace{1cm} (6)

And, according to equations (2) and (3) and taking into account the independence of the magnetic scattering cross-section upon $x$, one obtains

$$\frac{d\Sigma_m}{d\Omega}(Q) = \frac{d\Sigma}{d\Omega}(Q) \bigg|_{x=x_0}$$  \hspace{1cm} (7)

Consequently, in virtue of equations (1)-(3), within the Guinier’s scattering range, one may consider that for any value of $x$

$$\frac{d\Sigma_n}{d\Omega}(Q) = \frac{d\Sigma}{d\Omega}(Q) - \frac{d\Sigma_m}{d\Omega}(Q) \bigg|_{x=x_0}$$  \hspace{1cm} (8)

Equation (8) allows separating from experimental data the nuclear component of the scattering cross-section. Further, by plotting $\ln[\frac{d\Sigma_n}{d\Omega}(Q)]$ versus $Q^2$, one can establish the limits of the Guinier’s range for every value of $x$ (Table 1).
Table 1. The limits of the Guinier’s range for every value of $x$

| $X$  | $Q^2_{\text{min}}$(nm$^2$) | $Q^2_{\text{max}}$(nm$^2$) |
|------|-----------------|-----------------|
| 0.0  | 0.02272         | 0.2218          |
| 0.2  | 0.02272         | 0.2218          |
| 0.4  | 0.02272         | 0.2218          |
| 0.6  | 0.02272         | 0.2218          |
| 0.8  | 0.02272         | 0.1247          |
| 1.0  | 0.02272         | 0.1571          |

Extrapolation of the Guinier’s plots to $Q = 0$ gives $\frac{d\Sigma_n}{d\Omega}(0)$, and the square root of this, $[\frac{d\Sigma_n}{d\Omega}(0)]^{1/2}$, is drawn in Figure 4 as a function of $x$. The straight line representing the linear fit obtained by least squares method, has the following equation:

$$[\frac{d\Sigma_n}{d\Omega}(0)]^{1/2} = 2.20(\pm0.08) - 3.60(\pm0.14)x$$

Figure 4. Representation of $[\frac{d\Sigma_n}{d\Omega}(0)]^{1/2}$ as a function of $x$, together with the best straight line fit. The change of sign follows from the equation (3); actually, the quantity $N_{\rho}^{1/2} \rho \ast V_d$ is shown in the figure. Its value vanishes at $x = x_0 = 0.6$. The errors are smaller than the dimensions of the dots (circles).
The linear dependence on \( x \) of \( \frac{d^2 \Sigma}{d \Omega} (0) \), on one hand, and the vanishing value of this quantity at \( x = x_0 = 0.6 \), on the other, allows confirming the validity of the experimental data interpretation method.

For the interpretation of experimental data, a central-symmetrical colloidal particle with an impenetrable core of magnetite surrounded by a surfactant layer is considered. The volume of the layer is \( V_s = (4\pi/3)\left((R_0 + \delta)^3 - R_0^3\right) = 1.19 \cdot 10^{10} \text{ cm}^3 \); \( \delta = 20 \text{ Å} \) is the length of an oleic acid molecule (Figure 5).

\[
V_s = (4\pi/3)\left((R_0 + \delta)^3 - R_0^3\right) = 1.19 \cdot 10^{10} \text{ cm}^3 \; ; \; \delta = 20 \text{ Å} \text{ is the length of an oleic acid molecule.}
\]

![Figure 5](image)

**Figure 5.** Diagram of the volume fraction distribution of the components of the outer shell on the surface of the particle; (a) \( \phi \), the volume fraction of the surfactant; (b) \( \gamma \), the volume fraction of the carrier penetrated inside the surfactant shell; (c) \( (1 - \phi - \gamma) \), the empty volume fraction inside the surfactant shell.

At the same time, is assumed a nonuniform structure of the surfactant shell. So, it may be penetrated by the carrier and/or partially unoccupied. Assuming uniform densities inside the volume occupied by magnetite, surfactant and carrier and introducing \( \phi \), the volume fraction (referred to \( V_s \)) of the surfactant and \( \gamma \), the volume fraction of the carrier, taking into account the definition of \( \rho^* \) from equation (5) and the value of \( \rho^* \) for \( x = x_0 = 0.6 \), \( \rho^* = 0 \), the following expression can be written:

\[
(\rho_{mn} - \rho_c)V_0 + [\phi \rho_s - (1 - \gamma)\rho_c]V_s = 0
\]

Replacing with the given values \( \rho_{mn} \), \( V_0 \) the particle volume calculated from EM analysis data and \( V_s \) and \( \rho_s(x_0) = 3.777 \cdot 10^{10} \text{ cm}^2 \) for \( x_0 = 0.6 \), and considering \( \phi \rho_s \) negligible small, \( \gamma \), the volume fraction of the carrier inside the surfactant shell is obtained:

\[
\gamma = 0.25 \pm 0.1
\]

Further the volume and radius of the colloidal particle extracted from the carrier \( V_d \) and \( R_d \) are obtained:

\[
V_d = V_0 + (1 - \gamma)V_s
\]

\[
R_d = (3V_d / 4\pi)^{1/3} = (73.9 \pm 0.3) \cdot 10^{-8} \text{ cm}
\]

The value of the magnetite particle radius will be:
\[ R_p = R_d - \delta = (5.39 \pm 0.03) \text{nm} \]

where \( \delta \) is the length of the \( C_{18}H_{32}O_2 \) molecule adsorbed on the particle surface.

The rather high error in \( \gamma \) is caused by the particle size polydispersity. It should be noted, on the other hand, that in the present case, due to the very small value of \( \rho \), it is not possible to distinguish, in the total volume of the surfactant layer, the volume fraction \( \phi \) of the surfactant and, correspondingly, the volume fraction \((1 - \phi - \gamma)\) of the empty space. However, theoretical methods of statistical mechanics (see, e.g., reference [1]) give \( \phi \approx 0.3 \).

From the expression of the magnetic component of the differential scattering cross-section for \( Q = 0 \) and the value of \( d\Sigma(0)/d\Omega = 0.635 \text{cm}^{-1} \), the magnetic volume \( V_m \) and magnetic radius \( R_m \) may be inferred:

\[ R_m = \left( \frac{3V_m}{4\pi} \right)^{1/3} = (4.32 \pm 0.05) \text{nm} \]

These estimations indicate the existence of a nonmagnetic layer of thickness \( t_{nm} = 1 \text{nm} \) at the surface of the magnetite particle. Similar results have been reported in references [41,59].

4. Conclusions

Finally, from the results presented above, some conclusions may be drawn.

Within the Guinier scattering range, where the magnetic form factor, \( f \), of the magnetic nanoparticle is approximately 1 with an error less than 1%, using the contrast variation method when nuclear contrast changes the sign inside the concentration variation range, the contribution of nuclear and magnetic components of the experimental intensity can be established separately even in the case of unpolarised neutrons. This result, i.e. that these components can be distinguished with a contrast variation small angle neutron scattering, method, was first time reported in 90’s in the following papers [14-18] on SANS data from experiments accomplished at MURN instrument functioning at IBR-2 high pulsed reactor.

The values of physical and magnetic dimensions of the magnetic nanoparticles (from the same type of ferrofluids) obtained with the method presented above where confirmed lately by EM, magnetic measurements, SANS-POL and muon spectroscopy analysis and reported in several papers [44,61-65].

The surfactant layer has a rather complex structure. So, it is only partially occupied and may be penetrated by the carrier liquid. However, the negative coherent scattering length of the hydrogen leads to a relative small overall scattering length of the surfactant (which is mostly hydrogenated material). In some cases, as in the present one, this value is extremely small hindering the determination of the surfactant volume fraction. A solution of this problem in further SANS contrast variation investigations could be the deuteration of the surfactant.

The unavoidable size polydispersity of particles causes rather high errors in the determination of surfactant, carrier and empty space volume fractions in the outer shell of the magnetite core.

The results are compatible to the models where is supposed that the particle surface has a nonmagnetic layer. This might be caused accordingly to reference [59], by magnetite-surfactant interactions. In latest publications, however, such behaviour is treated as an intrinsic property of nonstructured magnetic materials (see, e.g., reference [60]).

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