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MAGNETIC SYSTEM FOR SMALL-ANGLE NEUTRON SCATTERING INVESTIGATION AT YUMO INSTRUMENT OF NANOMATERIALS

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Магнитная система для исследования наноматериалов малоугловым методом рассеяния нейтронов на спектрометре ЮМО

Измерения с помощью малоуглового рассеяния неполяризованных нейтронов (МУРН) могут дать численную информацию о магнитной микроструктуре, величине и микроструктуре магнитной анизотропии наноматериалов. Описана новая магнитная установка для малоуглового спектрометра ЮМО. Установка включает 2,5-Тл электромагнит, смонтированный на двухосном гониометрическом столе, источник питания, систему охлаждения, блок управления на персональном компьютере. Основными чертами магнитной системы являются: большой зазор для образцов (до 130 мм), автоматизированное вращение магнита в горизонтальной и вертикальной плоскости и большое пространство для держателя образцов. Система разработана в сотрудничестве с INCDIE ICPE-CA (Бухарест) и CIPEC SRL (Бухарест). Представлены первые экспериментальные результаты использования МУРН в магнитных жидкостях и магнитных эластомерах, полученные на спектрометре ЮМО, оборудованном новой магнитной системой.

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Magnetic System for Small-Angle Neutron Scattering Investigation at YUMO Instrument of Nanomaterials

SANS measurements using unpolarized neutron beams are able to provide quantitative information on the magnetic microstructure and the magnitude and microstructure of the magnetic anisotropy of nanomagnetic materials. Here we describe a new magnetic system for SANS at YUMO spectrometer. The system includes 2.5 T electromagnet established on a two-axes goniometric table, power supply, cooling system, PC-based control equipment. Main features of magnetic system are: big changeable gap for the samples (up to 130 mm size), computer controlled horizontal and vertical rotation and sufficiently large space for the sample holders. The system has been developed in cooperation with the INCDIE ICPE-CA (Bucharest) and CIPEC SRL (Bucharest). First experimental results of SANS in ferrofluids and magnetic elastomers obtained at YUMO spectrometer equipped with the new magnetic system are presented.

The investigation has been performed at the Frank Laboratory of Neutron Physics, JINR.

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INTRODUCTION

Neutron beams produced by different reactors are used for fundamental research on the structure and dynamics of the matter. Research efforts seek a better understanding of phase transitions, crystal structures, magnetic properties, superconductors, quantum liquids, fundamental properties of new materials.

Neutron scattering facilities have unique state-of-the-art capabilities for investigating structures and excitations of solid state matter for research in solid state physics, polymer science, and biology. The small-angle neutron diffractometer is widely used by colloid, polymer, biology scientists, scientists involved in investigation of nanomaterials and nanocomposites.

The instrument YUMO (Fig. 1) is located on the beamline *4* of the high pulsed IBR-2 reactor [1]. The useful wavelength range is $0.7 < \lambda/\text{Å} < 8$, the range of momentum transfers is $0.007 < Q/\text{Å}^{-1} < 0.5$. The time averaged neutron flux at the sample is up to $4 \cdot 10^7$ cm^{-2}s^{-1}.

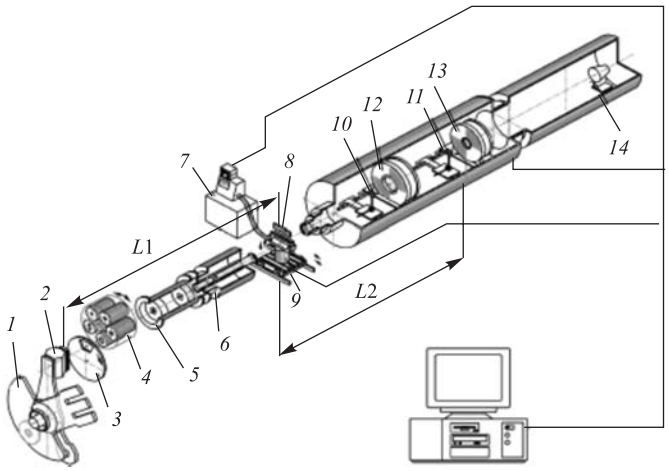

Fig. 1. YUMO SANS TOF spectrometer in function at IBR-2 reactor [1]: *1* — movable reflector; *2* — moderator/cold moderator; *3* — chopper; *4* — first collimator; *5* — vacuum tube; *6* — second collimator; *7* — liquid bath thermostat; *8, 9* — place for the electromagnet during SANS experiments on magnetic samples; *10, 11* — V, graphite, H_2O standards; *12* — circular detector of thermal neutrons; *13* — PSD circular detector of thermal neutrons; *14* — detector of direct beam

A variety of auxiliary devices is necessary for studies of materials in special environment conditions including low and high temperature, high pressure and strong magnetic fields.

For the investigation of magnetic nanostructures as nanoparticles, ferrofluids, magnetic gels and polymers, nanocarbon composites with magnetic properties, it is highly necessary during the experiment to apply magnetic fields of different intensity and orientation to the samples [2–11].

In the present paper a new magnetic system specially created for the YUMO SANS instrument is presented. The first measurements of SANS on magnetic elastomers and ferrofluids in applied magnetic field performed at YUMO diffractometer are given.

1. MAGNETIC SYSTEM AT YUMO SANS INSTRUMENT

The new magnetic system constructed for YUMO instrument is presented (Fig. 2). The magnetic system components are: the electromagnet with the goniometer, the power supply, computer for the automated command and control of the system.

The system was developed and constructed by the INCDIE ICPE-CA (Bucharest) in collaboration with CIPEC SRL (Bucharest).

The realization of the new position sensitive detector will permit the visualization and investigation of the magnetic field induced anisotropy in the analyzed sample.

Fig. 2. Magnetic system for YUMO SANS instrument

1.1. Technical Description. The electromagnet is a rigid system, made of two coils, with distilled water cooling system and two magnetic poles. The pole gap is variable, from 25 to 100 mm. On the both lateral parts of the electromagnet there is one slit, 100 × 600 mm, for giving the possibility to introduce the neutron flux. The whole electromagnetic system is sitting on one rotating system. There are two rotating planes with a rotation liberty of ± 40° in the vertical plane and of ± 95° in the horizontal plane. The rotating devices are equipped with electrical limiters, which stop the rotation motors. The motors are step-by-step motors. The positioning accuracy for both rotation planes is 0.1°.

The assembly electromagnet and rotating system are placed on a mobile support (translation).

The highest point of this support is adjustable, with ± 30 mm, for having the facility to the center of electromagnet in the neutron flux.

The electromagnet cooling system is a closed system, with distilled water. The cooling of the water is made by a water chiller wit with a capacity of 5000 W. The water flow is about 400 l/h.

The electromagnet is supplied by a DC source, with a current adjustment of 10 to 60 A DC. The adjustment accuracy is 0.7 % from the end of the scale. The magnetic field intensity is 2.5 T.

The electromagnet, with the turning system, is fixed on the translation support by means of some screws. The electromagnet can be picked up from the turning system with a crane. For picking up the ensemble electromagnet and turning system from the translation support, the electromagnet will be fixed to the turning system by means of some screws, before picking up.

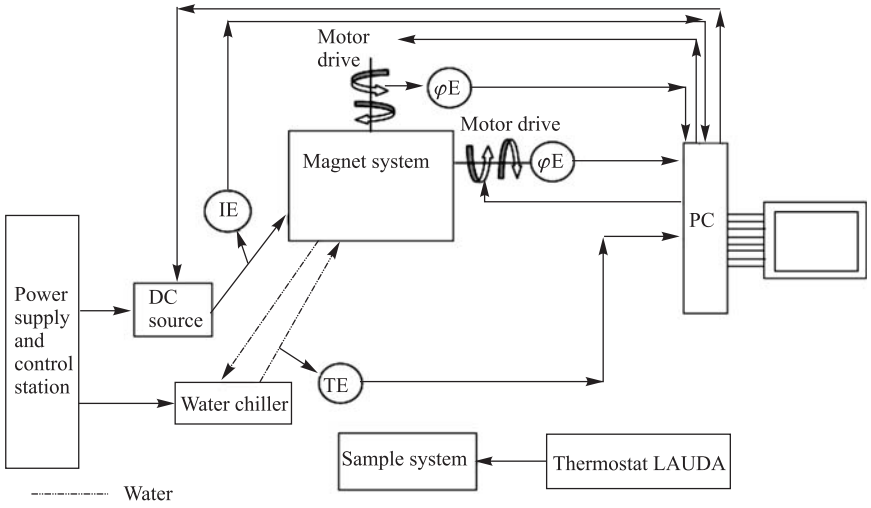

Fig. 3. Block diagram of the whole magnetic system

The electrical connections and those for water are elastical, giving the moving possibility to the electromagnet. These are very rapid connections.

The whole measuring and control system is connected with a PC. The software is a friendly one, who gives a lot of facilities:

1. Measuring and control for the following parameters:

— value of the magnetic field; value of current in the coils; cooling water temperature; water flow; electromagnet position in vertical plane; electromagnet position in horizontal plane; turning limits in vertical plane; turning limits in horizontal plane.

2. Alarms for outrunning of measuring values set points.

3. Curves diagrams for measuring values variations.

4. Historic of measuring values variations for a period of 30 days.

The technical drawings from Fig. 3 present the block diagram of the whole magnetic system.

2. FIRST MEASUREMENTS AND DATA ANALYSIS

2.1. Sample Preparation.

Magnetic Fluids. The sample was prepared at the Laboratory of Magnetic Fluids, CFATR, Romanian Academy, Timisoara Division, Timisoara, Romania [12, 13].

The sample is the magnetic fluid magnetite/oleic acid/d-benzene with volume fraction of magnetite of no more than $\varphi_m = 10\,\%$. The concentration of magnetite is determined from the value of the saturation magnetization (maximal magnetization of the sample) to be checked out before the standard measurements. The value of the sample magnetization corresponding to $\varphi_m = 10\,\%$ is 430 Gs in the external magnetic field $B > 1$ T. Magnetic particles are produced by the chemical condensation method as a result of the reaction:

$$\mathrm{FeCl_2 \cdot 4H_2O + 2FeCl_3 \cdot 6H_2O + 8NH_4OH \leftrightarrow Fe_3O_4 \downarrow + 8NH_4Cl + 20H_2O}. \tag{1}$$

The particle size distribution is described by the lognormal law with the maximally probable size $D_0 = 8 \pm 1$ nm and natural logarithm mean squared deviation $S = 0.3$. After the peptization, free surfactant (oleic acid), which is not absorbed on the magnetite surface, is removed from the system by a special chemical procedure. The ratio between contents of magnetite and surfactant in the final sample is about 1:1 (Fig. 4). Deuterated benzene is used as the carrier to analyze nuclear and magnetic scattering from the sample with good precision. The standard sample is highly stable (years) in closed containers and reproducible from the point of view of the preparation procedure.

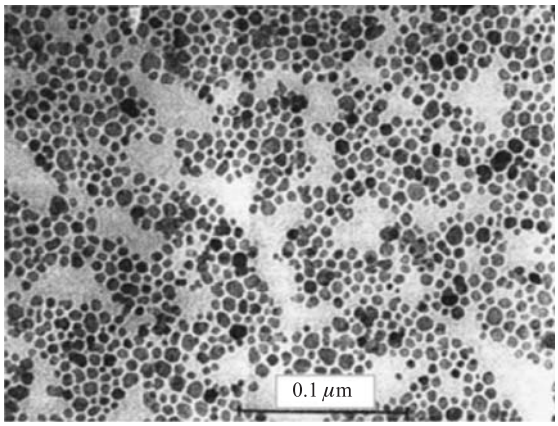

Fig. 4. TEM image of a magnetic fluid sample

Magnetic Elastomers. The studied samples, obtained at the Department of Electricity and Magnetism, West University of Timisoara [14–16], are composed of oil-based 7.7 % particle volume concentration Fe_3O_4 ferrofluid with oleic acid as surfactant, embedded in a polymer matrix formed from dimethylsiloxane, dibutyltindilaurate benzyl silicate.

2.2. Theoretical Background and Experimental Results. During standard measurements the sample in closed quarts cuvette is put under external magnetic field ($B > 1$ T) so that the saturation magnetization in the sample takes place. Then, the sample is irradiated by the thermal neutron beam followed by regis-

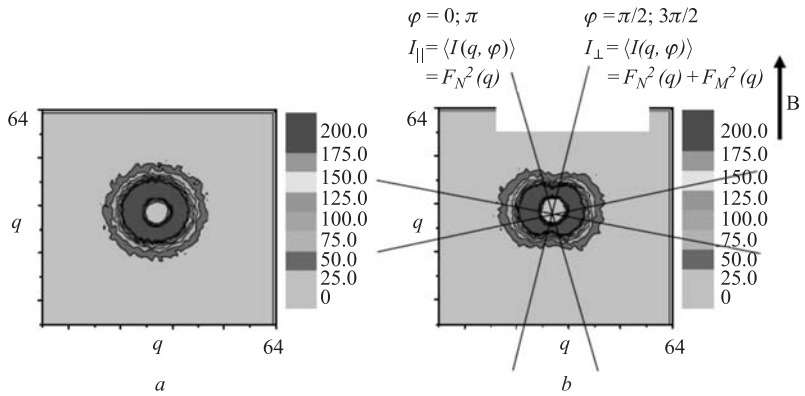

Fig. 5. The 2-dimensional scattering patterns from magnetic sample in absence (*a*) and presence (*b*) of external magnetic field. The separation of the nuclear and magnetic scattering contribution for saturated sample is demonstrated

tering of neutrons scattered at small angles (< 0.1 rad) by the position sensitive detector behind the sample. The 2-dimensional scattering pattern on the detector has specific features determined by two contributions: nuclear and magnetic scattering. To separate these contributions, two ways are used: (i) using nonpolarized neutron beam, and (ii) polarized neutrons.

The first way is for the nonpolarized neutron beam. In this case for the saturated sample the scattering intensity is

$$I(q,\varphi) = F_{N^2}(q) + F_{M^2}(q)\sin^2\varphi, \tag{2}$$

where $q = (4\pi/\lambda)\sin(\theta/2)$ is the module of the scattering vector with neutron wavelength, λ, and scattering angle, θ; φ is the radial angle on the detector from the zero-direction corresponding to direction of the strength of the applied magnetic field; $F_{N^2}(q)$ and $F_{M^2}(q)$ are the nuclear and magnetic scattering contributions, respectively. The separation is made by averaging of the scattering pattern for given q-value over radial angle φ around the vicinities of the directions parallel and perpendicular to the magnetic field (Fig. 5). This procedure transforms the 2-dimensional pattern to two 1-dimensional scattering curves:

$$I_{||} = \langle I(q,\varphi)\rangle_{\varphi=0;\pi} = F_{N^2}(q), \tag{3a}$$

$$I_{\perp} = \langle I(q,\varphi)\rangle_{\varphi=\pi/2;3\pi/2} = F_{N^2}(q) + F_{M^2}(q). \tag{3b}$$

These equations are the base to obtain nuclear and magnetic scattering contributions.

If the applied magnetic field does not produce the saturation magnetization in the sample the scattering intensity on the detector has the general form:

$$I(q,\varphi) = A(q) + B(q)\sin^2\varphi, \tag{4}$$

where $A(q)$ and $B(q)$ are functions corresponding to isotropic and anisotropic contributions to the scattering. The $B(q)$ function is determined purely by magnetic scattering of the partially oriented magnetic moments in the system. The $A(q)$ function is composed of isotropic nuclear scattering and isotropic part of magnetic scattering. The separation of these functions is made by averaging of the scattering pattern for given q-value over radial angle φ around the vicinities of the directions parallel and perpendicular to the magnetic field (Fig. 5). This procedure transforms the 2-dimensional pattern to two 1-dimensional scattering curves:

$$I_{||} = \langle I(q,\varphi)\rangle_{\varphi=0;\pi} = A(q), \tag{5a}$$

$$I_{\perp} = \langle I(q,\varphi)\rangle_{\varphi=\pi/2;3\pi/2} = A(q) + B(q). \tag{5b}$$

These equations are the base to obtain $A(q)$ and $B(q)$. Analysis of the magnetic system based on changes in these functions with the strength of the applied magnetic field is possible. The behavior of the standard sample with the change of the applied magnetic field is known [17].

3. TESTING FIRST RESULTS AT YUMO DIFFRACTOMETER WITH MAGNETIC FIELD

First measurements of SANS on magnetic elastomers and ferrofluids in applied magnetic field performed at YUMO diffractometer are presented [18]. Images (Fig. 6, *a–e*) from the new position sensitive detector [19] were obtained. The images present the summarizing experimental raw data for the wavelength of

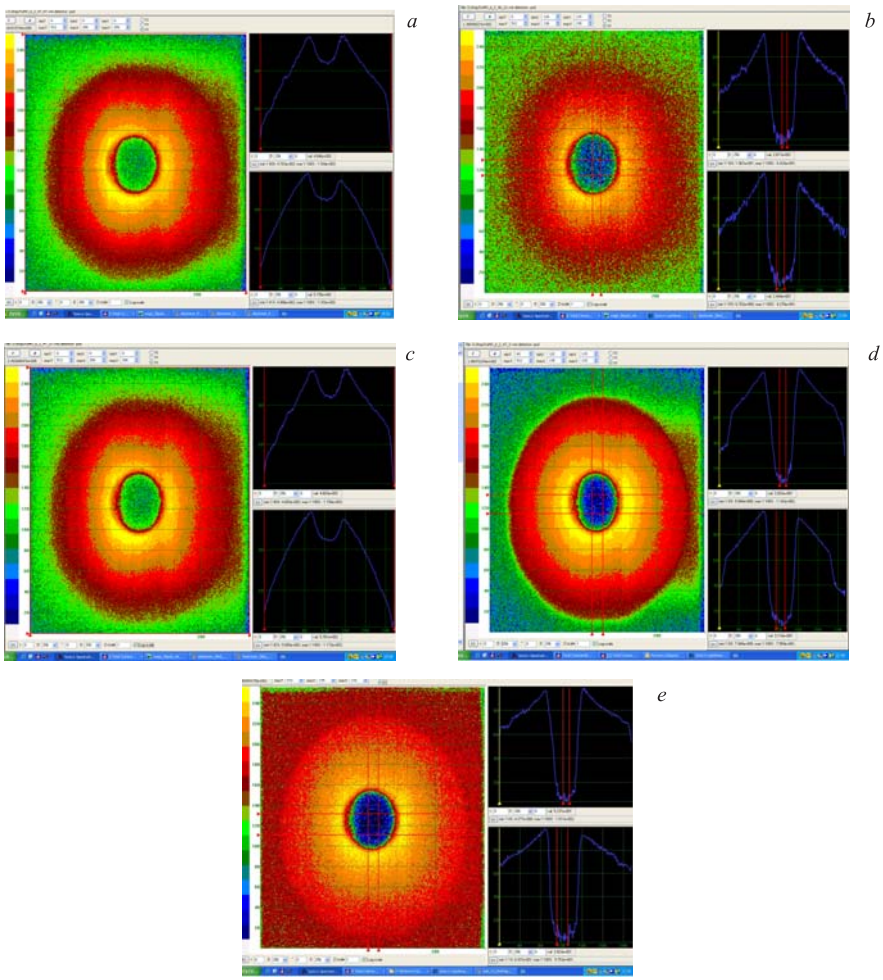

Fig. 6. The 2-dimensional scattering patterns from magnetic elastomer with Fe_3O_4 particles in magnetic field of 14 KOe (*a*), 10 KOe (*b*) and without magnetic field (*c*), and from ferrofluid with Fe_3O_4 particles in magnetic field of 1 KOe (*d*) and without magnetic field (*e*)

0.7–7 Å (in the left part of pictures). The profile of the space data at horizontal and vertical section is presented at the right side of the picture.

CONCLUSIONS

First experiments have demonstrated that the electromagnet together with the position sensitive detector is suitable for SANS measurements at YUMO instrument.

Further, a data analysis to transform the 2-dimensional pattern to 1-dimensional scattering curves as presented above in the formulae from (2) to (5) will be performed. For this task a new SAS program with a 2-dimensional analysis must be created.

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REFERENCES

1. *Kuklin A. I., Islamov A. Kh., Gordeliy V. I.* // Neutron News. 2005. V. 16, No. 3. P. 16–18.
2. *Kohlbrecher J., Wagner W.* // J. Appl. Cryst. 2000. V. 33. P. 804–806.
3. *Klose F., Ehlers G.* // Microscopy and Microanalysis. 2002. V. 8. P. 374–375.
4. *Weissmueller J. et al.* // Phys. Rev. B. 2001. V. 63. P. 214414.
5. *Wiedenmann A.* // Physica B. 2001. V. 297. P. 226.
6. *Yusuf S. M. et al.* // Phys. Rev. B. 2006. V. 74. P. 224428-1-11.
7. *Grabcev B. et al.* // J. Magn. Magn. Mater. 1999. V. 201. P. 140–143.
8. *Avdeev M. V. et al.* // J. Magn. Magn. Mater. 2004. V. 270. P. 371–379.
9. *Balasoiu M. et al.* // Magnetohydrodynamics. 2004. V. 40, No. 4. P. 359–368.
10. *Balasoiu M. et al.* // Romanian Reports in Physics. 2006. V. 58, No. 3. P. 305–313.
11. *Balasoiu M. et al.* // J. of Optoelectronics and Advanced Materials. 2008. V. 10, No. 11. P. 2932–2935.
12. *Bica D.* // Romanian Reports in Physics. 1995. V. 47, Nos. 3–5. P. 265–272.
13. *Bica D. et al.* // J. Magn. Magn. Mater. 2007. V. 311. P. 17–21.

14. *Bica I.* // J. Ind. Eng. Chem. 2006. V. 12, No. 5. P. 806–810.

15. *Bica I.* // J. Magn. Magn. Mater. 2002. V. 241. P. 196.

16. *Bica I.* // J. Ind. Eng. Chem. 2007. V. 11, No. 2. P. 299–304.

17. *Balasoiu M. et al.* // Experimental Report for Contract No. 08626319/021454-72: Realization of Standard Samples for the Test Measurements Necessary for Putting into Operation of the «Automated Installation for Spectrometer YUMO for Research of a Magnetic Field Influence on New Materials».

18. *Balasoiu M. et al.* // The 4th European Conference on Neutron Scattering 2007, Lundt, Book of Abstracts, Poster W072. P. 484.

19. *Kuklin A. I. et al.* // Ibid. Poster W118. P. 530.

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