Magnetic Sample Environment for the Small-Angle Neutron Scattering Spectrometer at CARR

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Abstract

Small Angle Neutron Scattering (SANS) instruments equipped with a magnetic sample environment can quantitatively analyze the magnetic microstructure of samples using unpolarized neutron beams. This paper reports on the composition, characteristics, and first experiments of a magnetic sample environment device installed on the Small Angle Neutron Scattering instrument (SANS) in the guide hall of the CARR reactor. The magnetic sample environment device mainly consists of a 3.00 T electromagnet, high-precision power supply, support table, cooling system, and microcomputer control system. The main features of the magnetic sample environment device are: uniform magnetic field; adjustable height of the electromagnet support table; adjustable air gap at the sample position within 120 mm; rotatable sample holder with adjustable height. Finally, this paper presents the experimental results of measuring nano-precipitates in 17-4PH stainless steel under the magnetic sample environment on the SANS instrument at the CARR reactor. These experimental results demonstrate that this magnetic sample environment device operates normally and meets the conditions for opening to SANS users.

Full Text

Preamble

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Magnetic Sample Environment Device for Small-Angle Neutron Scattering Instrument at CARR

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Abstract

Background: Small-angle neutron scattering (SANS) measurements using unpolarized neutron beams can quantitatively analyze the magnetic microstructure of samples. **Purpose:** To meet the demands of cutting-edge research, a magnetic sample environment device was developed for the SANS instrument at the CARR guide hall. **Methods:** The device consists of a 3.00 T electromagnet, high-precision power supply, support table, cooling system, and microcomputer control system. Key features include a uniform magnetic field, adjustable support table height, variable sample gap up to 120 mm, and a rotatable, height-adjustable sample holder. **Results:** Experimental results from measuring nanoscale precipitates in 17-4PH stainless steel under magnetic field conditions at the CARR SANS instrument are presented. **Conclusions:** These results demonstrate that the magnetic sample environment device operates normally and meets the requirements for user access.

Keywords: Neutron scattering, SANS, sample environment device, electromagnet

Introduction

Neutron beams produced by various neutron sources are utilized for fundamental research on material structure and dynamics, enabling better understanding of crystal structures, magnetic structures, phase transitions, magnetic properties, superconductivity, quantum liquids, and the basic characteristics of new materials [1-3]. Advanced neutron scattering facilities with sophisticated capabilities are employed to investigate material structure and excitations, finding unique applications in solid-state physics, polymer science, and biological research. Small-angle neutron scattering is a powerful technique for studying material structure and dynamic properties, particularly for organic materials such as molecular aggregates, polymers, and biological substances, offering irreplaceable capabilities unmatched by other methods [4-11].

The SANS instrument located at the end of the cold neutron guide (CNGD) at CARR [3] (shown in [Figure 1: see original paper]) utilizes a 50 mm × 50 mm section from the top of the CNGD as the neutron source. A neutron velocity selector provides wavelengths ranging from 4.0 to 20.0 Å with wavelength resolution between 10% and 22%. Sample sizes range from Φ5 to Φ25 mm, covering a scattering vector range of 0.0008 to 0.7 $Å^{-1}$. The instrument is equipped with a ^{3}He two-dimensional position-sensitive detector with an active area of 645 mm × 645 mm and a resolution of 5 mm × 5 mm.

Materials research typically requires various auxiliary equipment, particularly special sample environment devices for low temperature, high temperature, high pressure, and strong magnetic fields [5, 8-10, 12]. To investigate magnetic nanostructures such as magnetic nanocarbides, nanoparticles, magnetic fluids, magnetic gels, and polymers in nuclear-grade steels, applying magnetic fields of different strengths and orientations during experiments is essential [13, 14].

The magnetic sample environment device on the CARR SANS instrument is now operational. This system includes a 3.23 T electromagnet (factory setting) mounted on a height-adjustable support table, a high-precision power supply, cooling system, a sample holder with adjustable height and orientation, and a microcomputer-based control system. The primary characteristics of this magnetic sample environment device are: uniform magnetic field (magnetic field fluctuation less than 1% within $20\times20\times20 mm^{3}$ at 1 T), large variable gap at the sample position (up to 120 mm), and adjustable orientation and height of the sample holder.

This paper describes a magnetic sample environment device specifically developed for the CARR SANS instrument and presents the first experimental results from SANS measurements on a nuclear material (17-4PH stainless steel) under applied magnetic fields.

[Figure 1: see original paper] (A) Schematic diagram and (B) photograph of the CARR SANS instrument. (1) Pre-selector guide, (2) velocity selector shielding, (3) mechanical velocity selector, (4) attenuator, (5) third shutter, (6) post-selector guide, (7) collimator, (8) sample table and sample chamber, (9) gate valve, (10) two-dimensional position-sensitive detector, and (11) scattering vessel.

1 Magnetic Sample Environment Device for CARR SANS Instrument

This section describes the significance of the magnetic sample environment and details the construction and main technical specifications of the device developed for the CARR SANS instrument.

1.1 Significance of Magnetic Sample Environment

Magnetic field, like temperature and pressure, is a critical factor influencing material states and properties [5, 8-10, 12, 13]. Applied magnetic fields affect not only ferromagnetic and ferrimagnetic materials but also paramagnetic and diamagnetic materials.

Textured or anisotropic magnetic materials often exhibit superior performance in practical applications, making control of their microstructure crucial. Magnetic field treatment is a mature technique for enhancing texture and magnetic properties in magnetic materials. Some bulk materials consist of nanostructures with ordered crystalline arrangements, showing pronounced anisotropy and broad application potential. Research has shown that magnetic fields can cause inclusion migration in melts and induce grain alignment during preparation of both magnetic and non-magnetic materials.

In materials science, using strong magnetic fields to control materials during synthesis, chemical reactions, or crystal growth represents an effective method for creating and investigating novel materials. In-situ SANS studies of new phenomena induced by applied magnetic fields are being actively pursued in research on advanced materials such as magnetic materials, oxide superconductors, and organic compounds, as well as in investigations of microstructures in nanomaterials, spintronics, polymers, and biochemistry.

Another important aspect of using magnetic fields in SANS experiments is the ability to separately determine nuclear and magnetic scattering contributions. When studying magnetic materials with unpolarized neutron SANS, measuring scattered neutron intensity both without magnetic field and under a saturating magnetic field perpendicular to the scattering vector enables discrimination between nuclear and magnetic scattering contributions. Previously, at the CARR SANS instrument, such separation was only achievable for liquid samples using contrast variation methods [7, 11]. Magnetic neutron scattering under applied fields plays a central role in determining and understanding the microscopic properties of various magnetic systems, from establishing fundamental properties of magnetically ordered materials to elucidating the nature of magnetic characteristics in applications.

Magnetic fields that induce sample changes, such as material anisotropy and magnetic domain orientation, are typically on the order of several Tesla. The magnetic system on the CARR SANS instrument, shown in [Figure 2: see original paper], consists of an electromagnet with support table, a power supply with microcomputer control system, and a cooling system with automatic flow control. This magnetic sample environment device was designed in-house and developed with assistance from commercial companies.

[Figure 2: see original paper] Photograph of the magnetic system for the CARR-SANS instrument (A) cooling system, (B) power supply system, and (C) electromagnet fixed on support table).

1.2 Technical Description

The electromagnet is a robust device consisting primarily of a yoke, two coil sets, two pole columns, two pole tips, and a microcomputer-controlled power supply, equipped with a distilled water cooling system. The pole gap is adjustable from 0.0 to 120.0 mm.

The assembled electromagnet is mounted on a movable support table with adjustable height, ensuring the magnetic field center can be positioned between 1400 and 1550 mm above ground level to align with the neutron beam center. The electromagnet is powered by a DC supply with adjustable current from 0.0 to 65.0 A and a current regulation precision of 0.5% of the final value. The system provides a maximum magnetic field strength of 3.23 T and can maintain a stable 3.00 T field for extended periods. The cooling system is a closed-loop distilled water system with microcomputer-controlled flow, powered by a 7200 W chiller unit with maximum flow rate of approximately 1200 L/h. The microcomputer regulates flow based on set temperature.

The measurement and control system uses microcomputer control with a touch-screen interface and user-friendly software: (1) It measures and controls magnetic field value, coil current, cooling water temperature, and water flow rate. (2) It triggers alarms when measured values exceed setpoints. (3) It displays curves and charts of parameter variations.

[Figure 3: see original paper] shows the technical drawing and block diagram of the entire magnetic system.

[Figure 3: see original paper] Block diagram of the whole magnetic system.

2 Experimental Results and Analysis

This section describes the sample origin, standard measurement methods for SANS under applied magnetic fields, theoretical background for data analysis, and the first experimental results from the CARR SANS instrument with magnetic field capability.

2.1 Sample and Measurement Method

The experimental sample was extracted from a valve stem that served for 14 years in the main steam system of a nuclear power plant. The stem material is 17-4PH stainless steel [13]. Small pieces approximately 1 mm thick and 10 mm × 10 mm in size were cut and mounted in a sample support frame (shown in [Figure 4: see original paper]C) for SANS measurements under applied magnetic fields.

During measurement, the 17-4PH stainless steel sample was fixed at the center of an external magnetic field (B = 0.85 T) to achieve saturation magnetization. The sample was then irradiated with a collimated, monochromatic cold neutron beam. The scattering neutron patterns collected by the two-dimensional position-sensitive detector exhibit specific features determined by combined contributions from nuclear and magnetic scattering.

2.2 Theoretical Background for Data Analysis

For unpolarized neutron beams, the scattered neutron intensity from a saturated magnetized sample is given by:

$I(Q, \phi) = I\_N(Q) + I\_M(Q)\ \sin^2\phi$

where $Q = (4\pi/\lambda)\sin(\ /2)$ is the scattering vector for monochromatic neutrons of wavelength λ and scattering angle ; ϕ is the azimuthal angle on the detector measured from the direction of the applied magnetic field. I_N(Q) and I_M(Q) represent the contributions from nuclear and magnetic scattering, respectively. This separation is achieved by averaging the scattering patterns over azimuthal angles ϕ near directions parallel and perpendicular to the magnetic field for a given Q value. Software converts the two-dimensional patterns into two one-dimensional scattering curves. These equations form the basis for obtaining nuclear and magnetic scattering contributions.

If the applied magnetic field cannot fully saturate the sample, the scattering intensity on the detector is generally expressed as:

$I(Q, \phi) = A(Q) + B(Q)\sin^2\phi$

where A(Q) and B(Q) correspond to isotropic and anisotropic scattering contribution functions, respectively. The B(Q) function is determined entirely by magnetic scattering from partially aligned magnetic moments in the system. The A(Q) function comprises the isotropic nuclear scattering and the isotropic component of magnetic scattering. Separation of these functions is achieved by averaging scattering patterns over azimuthal angle ϕ near directions parallel and perpendicular to the magnetic field for a given Q value. Software converts the two-dimensional patterns into two one-dimensional scattering curves. These equations provide the foundation for obtaining A(Q) and B(Q). Analysis of magnetic systems based on the variation of these functions with applied magnetic field strength is possible because the magnetic field-dependent behavior of standard samples is well established.

2.3 Experimental Results

The results of CARR SANS measurements on 17-4PH stainless steel under magnetic sample environment conditions are shown in [Figure 4: see original paper]. Panels A and B present two-dimensional SANS patterns obtained from the two-dimensional position-sensitive detector, showing in-situ measurements under an applied magnetic field of 0.85 T and without magnetic field (0.00 T), respectively. These results are very similar to those obtained at the V4 instrument in Germany [13]. The data were processed and analyzed using BerSANS software [15], converting [Figure 4: see original paper]A into one-dimensional SANS curves parallel and perpendicular to the magnetic field direction, as shown in [Figure 4: see original paper]D.

[Figure 4: see original paper] Two-dimensional small-angle neutron scattering patterns from 17-4PH stainless steel sample: (A) in presence of external magnetic field, (B) in absence of external magnetic field, (C) sample holder, and (D) one-dimensional scattering curves parallel and perpendicular to the external magnetic field direction.

Conclusion

This paper reports on the magnetic sample environment device installed on the SANS instrument in the CARR guide hall, consisting primarily of a 3.00 T electromagnet, high-precision power supply, support table, cooling system, and microcomputer control system. Key features include uniform magnetic field, adjustable electromagnet support table height, variable sample gap up to 120 mm, and rotatable, height-adjustable sample holder. The standard measurement method for SANS under applied magnetic fields and its theoretical background are presented, along with the first experimental results from magnetic field experiments at CARR SANS. These results demonstrate that the magnetic sample environment device operates normally and meets the requirements for user access for magnetic small-angle neutron scattering experiments.

This device enables quantitative analysis of magnetic microstructures in samples and will contribute significantly to fundamental research and magnetic characterization of magnetic materials.

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References

[1] JIANG C. An overview of polarized neutron instruments and techniques in Asia Pacific [J]. AAPPS Bulletin, 2023, 33(1): 21.

[2] ANANIEV V, BELIAKOV A, BULAVIN M, et al. Pelletized cold moderator of the IBR-2 reactor: current status and future development [J]. Journal of Physics: Conference Series, 2016, 746(1): 012031.

[3] CHEN D F, LIU Y T, GOU C, et al. Development of neutron scattering on 60MW research reactor in CIAE [J]. Physica B, 2006, 385-386: 966-7.

[4] X. M. DU, H. T. QI, T. F. LI, et al. SMALL-ANGLE NEUTRON SCATTERING CHARACTERIZATION OF GRAPHENE/Al NANOCOMPOSITES [J]. Digest Journal of Nanomaterials and Biostructures, 2019, 14(2): 329-35.

[5] SAKAGUCHI Y, TAKATA S, ARIMA H, et al. High temperature furnace for small-angle neutron scattering instrument at J-PARC [J]. Journal of Neutron Research, 2019, 21(1-10).

[6] LI T-F, CHENG Y-Y, WANG Y, et al. Analysis of Dimer Impurity in Polyamidoamine Dendrimer Solutions by Small-angle Neutron Scattering [J]. Chinese Journal of Polymer Science, 2019, 37(8): 827-33.

[7] ZHANG H, HE C, YUAN G, et al. Thirty meters small angle neutron scattering instrument at China advanced research reactor [J]. Nuclear Instruments and Methods in Physics Research, 2014, 735(jan.): 490-5.

[8] KOHLBRECHER J, BOLLHALDER A, VAVRIN R, et al. A high pressure cell for small angle neutron scattering up to 500MPa in combination with light scattering to investigate liquid samples [J]. REVIEW OF SCIENTIFIC INSTRUMENTS, 2007, 78(12): 125101.

[9] KOMPATSCHER M, BäR M, HECHT J, et al. A high-temperature cell for in situ small-angle neutron scattering studies of phase separation in alloys [J]. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 2002, 495(1): 40-7.

[10] KERCH H M, BURDETTE H E, LONG G G. A High-Temperature Furnace for In Situ Small-Angle Neutron Scattering during Ceramic Processing [J]. J Appl Cryst 1995, 28(5): 604-10.

[11] HOLLAMBY M J. Practical applications of small-angle neutron scattering [J]. Physical Chemistry Chemical Physics, 2013, 15(26): 10566-79.

[12] KLIMKO S, ZHERNENKOV K, TOPERVERG B P, et al. Development and application of setup for ac magnetic field in neutron scattering experiments [J]. Review of Scientific Instruments, 2010, 81(10): 103303.

[13] 闫士博, 李天富, 刘蕴韬, 等. 17-4PH 不锈钢纳米析出物的磁小角中子散射研究 [J]. 原子能科学技术, 2022, 56(3).

[14] ZIJUN W, XIAOMING D, TIANFU L, et al. Investigation of Cu-enriched precipitates in thermally aged Fe-Cu and Fe-Cu-Ni model alloys [J]. Journal of Nuclear Materials, 2022, 562: 153601.

[15] KEIDERLING U. The new 'BerSANS-PC' software for reduction and treatment of small angle neutron scattering data [J]. Appl Phys A, 2002, 74(1): s1455-s7.

Note: Figure translations are in progress. See original paper for figures.