## **Experimental report**

Proposal:	5-32-8	16	<b>Council:</b> 10/2014					
Title:	Imagir	Imaging of displacement fields in a bulk amorphous alloy						
Research area: Physics								
This proposal is a new proposal								
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Samples: bulk amorphous alloy (metal)								
Instrument			Requested days	Allocated days	From	То		
D33			6	5	07/05/2015	12/05/2015		
Abstract								

We will investigate the magnetic microstructure of a bulk metallic glass (BMG) (Fe70Mo5Ni5P12.5B2.5C5) by means of spin-polarized and magnetic-field-dependent small-angle neutron scattering (SANS). Recent theoretical work predicts that the fundamental instability of a BMG - when subjected to a compressive stress - is accompanied by the appearance of coherent quadrupolar structures forming along a line which is at an angle of 45° relative to the compressive stress axis. As consequence of magnetoelastic coupling (linking the strain tensor to the magnetization), we expect that the displacement field related to the compressive stress couples to the magnetization distribution. SANS is ideally suited to image the resulting quadrupolar spin texture in the bulk of the material and on the nanometer length scale. The proposed POLARIS experiments will provide insights into the existence and structure of displacement fields in a BMG, and we may, thus, contribute to the understanding of the fundamental instability and to the improvement of the mechanical properties of this important class of materials.

## Influence of mechanical deformation on the magnetic neutron scattering of the bulk metallic glass Fe<sub>70</sub>Mo<sub>5</sub>Ni<sub>5</sub>P<sub>12.5</sub>B<sub>2.5</sub>C<sub>2.5</sub>

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Bulk metallic glasses (BMG) are amorphous solids which are well known for their excellent mechanical properties.<sup>1</sup> In this experimental report, we focus on their magnetic behavior and we discuss the influence of mechanical deformation on the magnetic microstructure of BMG  $Fe_{70}Mo_5Ni_5P_{12.5}B_{2.5}C_{2.5}$  as seen by magneticfield-dependent small-angle neutron scattering (SANS). We present and compare the results of unpolarized SANS measurements on various BMG samples (as-cast, aged, deformed), and we analyze the SANS cross sections in real space by computing the correlation function.

The SANS experiment was carried at the instrument D33 at the Institut Laue-Langevin (ILL), Grenoble, France. The measurements were made using unpolarized incident neutrons with a mean wavelength of  $\lambda = 6 \text{ Å}, \Delta \lambda / \lambda \sim 10\%$  (FWHM), and for a *q*-range of  $0.035 \text{ nm}^{-1} \leq q \leq 1.5 \text{ nm}^{-1}$ . The magnetic field was applied perpendicular to the incident neutron beam ( $\mathbf{k}_0 \perp \mathbf{H}$ ). Neutron data were recorded by first applying a large positive field (1496 mT), which is assumed to saturate the sample (compare Fig. 1), and then reducing the field following the magnetization curve. All data were collected at room temperature. SANS data reduction (correction for background scattering, transmission, detector efficiency) was carried out using the GRASP software package.

For the current experiment, amorphous BMG samples of nominal composition Fe<sub>70</sub>Mo<sub>5</sub>Ni<sub>5</sub>P<sub>12.5</sub>B<sub>2.5</sub>C<sub>2.5</sub> were produced at Glassimetal Technology Inc. From the ascast cylinder (diameter: 4 mm), three discs with thicknesses of, respectively: 0.36, 0.38, and 0.28 mm were cut. One sample remained in the as-cast state, one specimen was aged for 8 hours at a temperature of 631 K in nitrogen atmosphere, and one disc was mechanically deformed; the compressive force was applied in the plane of the disc with a maximum value of  $10^5$  N. As a result of the deformation, the diameter of the disc in the direction of the applied force was reduced from 4.0 mm to 3.6 mm, with no reduction of the diameter in the perpendicular direction. The amorphous character of the samples was continuously monitored by means of wide-angle X-ray diffraction. The magnetic properties of the samples were studied using a vibrating sample magnetometer (LakeShore VSM 7400). Magnetization data were recorded at room temperature for applied magnetic fields up to  $\pm 2 \mathrm{T}$ ; the applied-field direction was in the plane of the discs.

For the scattering geometry where the applied magnetic field (assumed to be parallel to the  $\mathbf{e}_z$  direction of a Cartesian laboratory coordinate system) is perpendicular to the incident neutrom beam ( $\mathbf{k}_0 \perp \mathbf{H}$ ), the elastic unpolarized SANS cross section  $d\Sigma/d\Omega$  at momentumtransfer vector  $\mathbf{q}$  can be written as<sup>2</sup>

$$\begin{split} \frac{d\Sigma}{d\Omega}(\mathbf{q}) &= \frac{8\pi^3}{V} \left( |\widetilde{N}|^2 + b_H^2 |\widetilde{M}_x|^2 + b_H^2 |\widetilde{M}_y|^2 \cos^2\theta + b_H^2 |\widetilde{M}_z|^2 \sin^2\theta - b_H^2 (\widetilde{M}_y \widetilde{M}_z^* + \widetilde{M}_y^* \widetilde{M}_z) \sin\theta \cos\theta \right), \end{split}$$

where  $\mathbf{q}$  is the scattering or momentum-transfer vector, V is the scattering volume,  $b_H = 2.91 \times 10^8 \,\mathrm{A^{-1}m^{-1}}$ relates the atomic magnetic moment to the Bohr magneton, and  $\widetilde{N}(\mathbf{q})$  and  $\widetilde{\mathbf{M}}(\mathbf{q}) = [\widetilde{M}_x(\mathbf{q}), \widetilde{M}_y(\mathbf{q}), \widetilde{M}_z(\mathbf{q})]$ denote, respectively, the Fourier coefficients of the nuclear scattering-length density and of the magnetization  $\mathbf{M}(\mathbf{r})$ ;  $\theta$  represents the angle between  $\mathbf{H}$  and  $\mathbf{q}$  so that  $\mathbf{q} \cong q[0, \sin \theta, \cos \theta]$ ; the asterisks "\*" mark the complex-conjugated quantity and the atomic magnetic form factor (in the expression for  $b_H$ ) is approximated to unity since we are dealing with forward scattering.

As shown in Ref. 3, near magnetic saturation and for a weakly inhomogeneous bulk ferromagnet,  $d\Sigma/d\Omega$  can be evaluated by means of micromagnetic theory. In particular, it follows that

$$\frac{d\Sigma}{d\Omega}(\mathbf{q}) = \frac{d\Sigma_{\rm res}}{d\Omega}(\mathbf{q}) + \frac{d\Sigma_M}{d\Omega}(\mathbf{q}),\tag{1}$$

where

$$\frac{d\Sigma_{\rm res}}{d\Omega}(\mathbf{q}) = \frac{8\pi^3}{V} \left( |\widetilde{N}|^2 + b_H^2 |\widetilde{M}_z|^2 \sin^2\theta \right)$$
(2)

represents the nuclear and magnetic residual SANS cross section, which is measured at complete magnetic saturation (infinite field), and the remaining part—the so-called spin-misalignment SANS cross section—can be written as

$$\frac{d\Sigma_M}{d\Omega}(\mathbf{q}) = S_H(\mathbf{q}) \, R_H(q,\theta,H_i) + S_M(\mathbf{q}) \, R_M(q,\theta,H_i).$$
(3)

 $d\Sigma_M/d\Omega$  contains the purely magnetic scattering due to transversal spin components, with related Fourier amplitudes  $\widetilde{M}_x(\mathbf{q})$  and  $\widetilde{M}_y(\mathbf{q})$ . This cross section further decomposes into a contribution  $S_H R_H$  due to perturbing magnetic anisotropy fields and a part  $S_M R_M$  related to

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FIG. 1. Room-temperature magnetization curves of as-cast, deformed, and aged BMG alloy  $Fe_{70}Mo_5Ni_5P_{12.5}B_{2.5}C_{2.5}$ . Inset zooms into the approach-to-saturation regime; dashed vertical lines indicate the field values used in the scattering experiment.

magnetostatic fields. The latter contribution is proportional to the jump  $\Delta M$  of the saturation magnetization on microstructural defects. We remind the reader that the micromagnetic SANS theory (Ref. 3) considers a uniform exchange interaction and a random distribution of magnetic easy axes, but takes explicitly into account spatial variations in the magnitude of the magnetization. In a simple model, the  $\widetilde{M}_z^2$  Fourier component can be expressed as

$$\widetilde{M}_{z}^{2}(q) = \frac{(\Delta M)^{2}}{(8\pi)^{3}} V_{p}^{2} P(q) S(q), \qquad (4)$$

where  $\Delta M$  is the average jump of the magnetization magnitude on the defect,  $V_p$  is the defect volume, P(q) its form factor, and S(q) denotes the structure factor.

The results of the magnetization measurements are shown in Fig. 1. All samples are classified as soft magnetic, and a field of about 0.2 T is sufficient to bring them into the magnetically saturated state. The highfield magnetization of the aged sample is slightly larger than the magnetization of the other two samples.

Figure 2 shows the field dependence of the neutron countrate. In the saturated state, the countrate is almost as low as the background scattering, which indicates an extremely weak residual scattering contribution (of the order of 1-2 counts/s) and suggests that the spin-misalignment scattering dominates the measured total  $d\Sigma/d\Omega$  at lower fields. Reducing the applied field below about 200 mT, results in a strong increase of the countrate, concomitant with the decrease of the overall average magnetization (compare Fig. 1). While for the as-cast and deformed samples we observe the saturation of the countrate for fields below about 50 mT, the countrate of the aged sample exhibits a maximum at about 80 mT.

The experimental two-dimensional unpolarized SANS cross sections  $d\Sigma/d\Omega$  at selected fields are depicted in Fig. 3. The scattering at saturation (1496 mT) is weak



FIG. 2. Field dependence of the neutron countrate of as-cast, deformed, and aged BMG alloy  $Fe_{70}Mo_5Ni_5P_{12.5}B_{2.5}C_{2.5}$ . Sample-to-detector position is 13.5 m, which corresponds to  $0.035 \text{ nm}^{-1} \le q \le 0.3 \text{ nm}^{-1}$ .

and isotropic for all samples investigated. By comparison to the expression for the residual SANS cross section [Eq. (2)], this observation implies that either the isotropic nuclear scattering ( $\propto |\tilde{N}|^2$ ) is very much larger than the anisotropic longitudinal magnetic scattering ( $\propto |\tilde{M}_z|^2 \sin^2 \theta$ ), or, if the nuclear SANS is smaller or comparable to the  $|\tilde{M}_z|^2 \sin^2 \theta$  term, that fluctuations in the magnitude of the saturation magnetization are negligible, because no  $\sin^2 \theta$  angular anisotropy is observed and  $|\tilde{M}_z|^2 \propto (\Delta M)^2$  [compare Eq. (4)]. Since, however, for this particular alloy the nuclear scattering lengthdensity contrast is almost equal to the magnetic contrast, it follows that spatial variations in the magnitude of the saturation magnetization are small in this material.

By decreasing the field down to the remanent state, the SANS pattern reveals a combination of different angular anisotropies which can be related to the different origins of magnetic scattering. In particular, we refer to the data at 70 mT which show a pronounced maximum of the neutron intensity for directions parallel and antiparallel to the applied field. This so-called "spike" anisotropy has previously been found in a SANS study on sintered Nd-Fe-B magnets.<sup>4</sup> It is related to the formation of flux-closure patterns on a nanometer length scale. It is also worth noting that the  $d\Sigma/d\Omega$  of  $Fe_{70}Mo_5Ni_5P_{12.5}B_{2.5}C_{2.5}$  shows (at all q and H investigated) no sign of the so-called "clover-leaf" anisotropy with maxima roughly along the diagonals of the detector; such an angular anisotropy would indicate the presence of inhomogeneities in the saturation magnetization. In other words, the absence of the "clover-leaf" anisotropy is a logical consequence of negligible  $|\widetilde{M}_z|^2$  contributions to  $d\Sigma/d\Omega$  (see the above the discussion of the 2D data in the saturated state). The field-dependent SANS data will be further analyzed in terms of our micromagnetic theory and by computing the correlation function of the spin misalignment (see Fig. 4).

 $\mu_0 H = 1496 \text{ mT}$  $\mu_0 H = 163 \text{ mT}$  $\mu_0 H = 109 \text{ mT}$  $\mu_0 H = 70 \text{ mT}$  $\mu_0 H = 44 \text{ mT}$  $\mu_0 H = 2 \text{ mT}$ As-cast  $\mu_0 H = 2 mT$  $\mu_0 H = 163 \text{ mT}$  $\mu_0 H = 109 \text{ mT}$  $\mu_0 H = 70 \text{ mT}$  $\mu_0 H = 44 \text{ mT}$  $\mu_0 H = 1496 \text{ mT}$ Aged  $\mu_0 H = 1496 \text{ mT}$  $\mu_0 H = 163 \text{ mT}$  $\mu_0 H = 109 \text{ mT}$  $\mu_0 H = 70 \text{ mT}$  $\mu_0 H = 44 \text{ mT}$  $\mu_0 H = 2 \text{ mT}$ Deformed

FIG. 3. Two-dimensional unpolarized SANS cross sections  $d\Sigma/d\Omega$  of as-cast, deformed, and aged BMG alloy  $Fe_{70}Mo_5Ni_5P_{12.5}B_{2.5}C_{2.5}$  at selected applied magnetic fields ( $\mathbf{k}_0 \perp \mathbf{H}$ ) (logarithmic color scale).



FIG. 4. 2D correlation functions of the spin-misalignment SANS cross section of as-cast, deformed, and aged BMG alloy  $Fe_{70}Mo_5Ni_5P_{12.5}B_{2.5}C_{2.5}$  at  $\mu_0H = 130$  mT.

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