Functional Materials 11, No.3 (2004)

Small-angle neutron scattering in Invar Fe–Ni–C alloys in magnetic field

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The small-angle neutron scattering (SANS) in Invar Fe–Ni and Fe–Ni–C alloys was studied using polarized neutrons. The measurements were carried out without and with applied magnetic field of 2.5 T at the sample perpendicular to the neutron beam. The nonlinear SANS curves were obtained within the range of scattering vector $q = 0.006–0.025$ Å^{-1} and analyzed using power law function. The considerable magnetic contribution to neutron scattering has been demonstrated. The size of aggregates has been estimated using the Indirect Fourier Transformation method.

Исследовано малоугловое рассеяние нейтронов (МУРН) в инварных Fe–Ni и Fe–Ni–C сплавах с использованием поляризованных нейтронов. Измерения проведены без магнитного поля и с наложенным к образцу магнитным полем 2,5 Т, перпендикулярным направлению пучка нейтронов. Получено нелинейную кривую МУРН в интервале векторов рассеяния $q = 0.006–0.025$ Å^{-1}, проанализировано с использованием степенной функции. Продемонстрирован существенный магнитный вклад в рассеяние нейтронов. С использованием косвенного фурье-преобразования оценены размеры неоднородностей.

The physical models explaining the Invar anomaly in the f.c.c. Fe — 36 % Ni alloy assume an essential magnetic contribution [1, 2]. As shown in [3, 4], the small-angle neutron scattering (SANS) in Invar alloys containing 30 to 35 % Ni has magnetic nature and the magnetic inhomogeneities give considerable contribution to SANS along with the magnon and critical neutron scattering at $(T_c - T)/T_c = 0.21$ (T_c is the Curie point). Carbon in Fe–Ni solid solution causes variations in the magnetic structure in a wide range and this effect depends on Ni concentration [5, 6]. SANS experiments in the Fe — 30.5 % Ni — 1.5 % C (wt.%) and Fe — 30.3 % Ni alloys have shown that solution of carbon increases the neutron scattering intensity and changes considerably the slope of the SANS curve [7]. Two values of the power-law exponent characterising different size and structure of inhomogeneities were obtained in [7]. A similar double-slope SANS curve was obtained for the Fe70Ni30 (0.1 at. % C) alloy [8] that was explained by existing of two correlation radii estimated from the temperature dependences of SANS amplitudes.

In this work, the SANS experiment was performed on the Invar type Fe–Ni and Fe–Ni–C alloys using polarized neutron beam and applied magnetic field in order to separate the nuclear and magnetic components to SANS and to support idea concerning the presence of magnetic and chemical inhomogeneities in Invar alloys.

The Fe — 30.3 % Ni and Fe — 30.5 % Ni — 1.5 % C (wt.%) alloys were melted in a vacuum induction furnace in protective argon atmosphere. The ingots were aged at 1000°C for 3 h. The carbon concentration was determined by chemical analysis and the nickel content was obtained using the X-ray fluorescence analysis. The samples were shaped as 2 to 2.5 mm thick plates treated at 1100°C in vacuum and subsequently quenched in water. X-ray phase analysis was used to control the phase composition of the alloys.

SANS experiments were performed using the SANS1 setup at the FRG1 research reactor at GKSS Research Centre, Geesthacht, Germany [9]. The neutron wavelength was 8.5 Å and the wavelength resolution was 10 % (full-width-at-half-maximum value). The range of scattering vectors ($0.005 < q < 0.025$ Å^{-1}) was obtained using four sample-to-detector distances (0.7 to 9.7 m). A polarized neutron beam was used in SANS experiments. The experiments were carried out at room temperature and in applied magnetic field of 2.5 T, on the samples being perpendicular to the neutron beam. The initial polarization of the neutrons (parallel to direction of the magnetic field) was close to 1, while the efficiency of the spin flipper to realize the inverse polarization state (antiparallel to direction of the magnetic filed) was 0.9.

The two-dimensional isotropic scattering spectra were corrected for the detector efficiency by dividing by the incoherent scattering spectra of pure water measured using a 1 mm path length quartz cell. The smearing induced by different instrumental settings was included in the data analysis. For each instrumental setting, the ideal model cross-section was disturbed by the appropriate resolution function when the model scattering intensity was compared to the measured one by means of least-squares methods. The parameters in the models were optimized by conventional least-squares analysis and the errors of the parameters were calculated by conventional methods [10].

The mathematical formalism connected with data treatment of SANS patterns for magnetic materials is described in [11]. Below, formulas for differential scattering cross-sections are presented assuming that the interparticle correlation is small and can be neglected.

In the case of polarized neutrons, an interference between nuclear ($F_N(q)$) and magnetic scattering ($F_M(q)$) takes place. When magnetic moments in the sample are oriented along the applied magnetic field direction, the scattering intensities depend on radial angle φ between directions of the applied magnetic field and scattering vector q; and on the polarization states of the incident neutron beam parallel (-) and antiparallel (+) to the direction of magnetic filed as:

$$I^-(q,\varphi,H) = \\ = F_N^2(q) + (F_M^2(q) + 2P\cdot\varepsilon F_N(q)F_M(q))\sin^2\varphi, \tag{1a}$$

$$I^+(q,\varphi,H) = \\ = F_N^2(q) + (F_M^2(q) - 2PF_N(q)F_M(q))\sin^2\varphi, \tag{1b}$$

where P is the polarization of the neutron beam, and ε, the efficiency of the spin-flipper. Averaging over φ gives:

$$I^-(q,H) = \\ = F_N^2(q) + (1/2)F_M^2(q) + P\cdot\varepsilon F_N(q)F_M(q), \tag{2a}$$

$$I^+(q,H) = \\ = F_N^2(q) + (1/2)F_M^2(q) - PF_N(q)F_M(q), \tag{2b}$$

2D patterns and φ-averaged 1D curves contain contributions from nuclear ($F_N{}^2(q)$) and magnetic ($F_M{}^2(q)$) scattering and also additional cross term $F_N(q)F_M(q)$. The summation of the scattering for both polarization states of the initial beam gives

$$I(q,H{=}2.5) = F_N^2(q) + (1/2)F_M^2(q). \tag{3}$$

In the absence of magnetic field, both the nuclear and magnetic scattering are isotropic over the radial angle φ and the scattering intensity comprises these two contributions in the following proportion:

$$I(q,H{=}0) = F_N^2(q) + (2/3)F_M^2(q). \tag{4}$$

From Eq. 1–4, it is possible to separate immediately nuclear and magnetic contributions out of the total scattering. From another point of view, the possible changes of magnetic structure under applied magnetic field should be checked before start the separation analysis. In Fig. 1, the φ-averaged scattering intensities in the Fe — 30.3 % Ni (wt. %) alloy for $H = 0$ and $H = 2.5$ T are presented. The intensity is observed to drop dramatically (by a factor of 5 to 20) when magnetic field is applied. This points that the applied magnetic field (2.5 T) significantly destroys (or at least changes) the magnetic structure in studied alloy.

The obtained decrease of scattering intensities by about one order makes it possible to consider the scattering at $H = 0$ to be very close to the scattering on $F_M(q)$ (magnetic structure of alloys) and the scattering at $H = 2.5$ T, to be very close to that on $F_N(q)$ (nuclear structure of alloys). We can support our suggestion by negligible difference between curves representing different spin directions $I^-(q,H)$ and $I^+(q,H)$ shown in Fig. 2. However, it should be noted that the magnetic scattering is still present at $H = 2.5$ T because we observed an anisotropy of 2D scattering pattern under applied field.

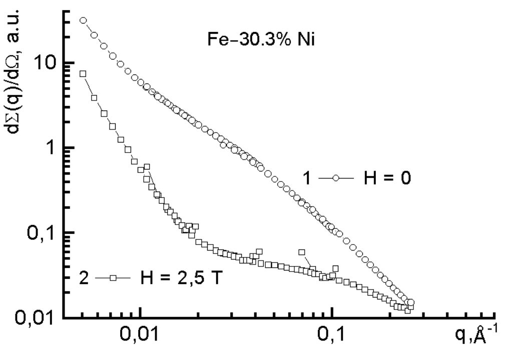

Fig. 1. Polarized neutrons SAS (averaged by φ-angle and summarized by spin direction) in the Fe — 30.3 % Ni (wt. %) alloy (for $H = 0$ and $H = 2.5$ T) after annealing of sample at 1100°C in vacuum for 30 min.

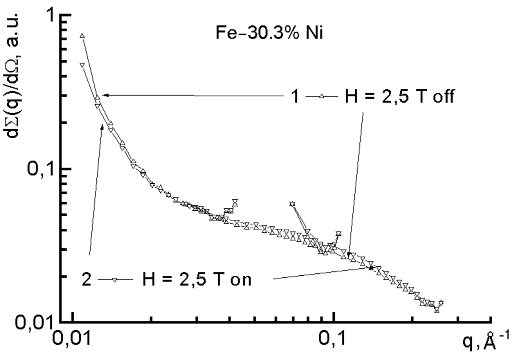

Fig. 2. SANS in the Fe — 30.3 % Ni (wt. %) alloy at $H = 2.5$ T by different polarization directions of neutron beam (averaged by φ angle).

To characterize the formed structures, the power law was applied for approximation of the SANS curves: $I(q) = Aq^{-\alpha} + B$, where α is the power index; A, the contrast factor, and B, the residual incoherent background. The estimations gave values of the power index at low and large scattering vectors q listed in Table. The α values point the kinds of structures which give the scattering in the different interval of scattering vector q. For the Fe — 30.3 % Ni alloy at $H = 0$, where we have assumed the most scattering on magnetic structure takes place, the α value is within limits of 1.6 to 2.3. This means the mass fractal structure $M \sim r^{\alpha}$, here the M (mass) corresponds to magnetic moments of magnetic structures and r is a linear size [12].

Under magnetic field, the scattering mostly on nuclear inhomogeneities is observed. At large length scale ($r > 200$Å), the scattering from surface fractal is observed with some substructure of smaller aggregates. To estimate the size of the smaller aggregates of nuclear structures in the Fe — 30.3 % Ni alloy, the data at $q > 0.05$ Å^{-1} were analysed using the Indirect Fourier Transformation (IFT) method [13] in the form as developed by Pedersen [14]. The Fourier transformation of the experimental curve provides a pair distance distribution function $p(r)$. The aggregate parameter, such as the radius of gyration, R_g, is calculated from $p(r)$ and is equal to 16±1 Å (the equivalent sphere radius is 22 Å).

The anisotropy of 2D scattering pattern for the Fe — 30.5 % Ni — 1.5 % C alloy is presented in Fig. 3 showing that the some magnetic scattering is still present even at $H = 2.5$ T, especially at large q. However, the applying of magnetic field decreases significantly the average scattering intensities (Fig. 4) that is why we have applied the same approach to interpret the scattering data, i.e., at $H = 0$, one observes scattering mostly from magnetic structures, while at $H = 2.5$ T, only from nuclear structures. The parameters of curves are presented in Table 1.

The scattering on magnetic structures at large scale length ($r > 300$ Å) follows the Porod's law (α is about 4), which corresponds to object with smooth and sharp surface [15]. At smaller length scales (100–300 Å), we observe the scattering on fractal-like aggregates (mass-fractal, because $\alpha = 2.6\pm0.1$) and with decreasing of size (down to 20 Å), one observes the scattering on fractal-like surface of these aggregates ($\alpha = 3.3\pm0.1$) [12]. The obtained results support data on fractal-like properties of aggregates derived in the Fe — 30.5 % Ni — 1.5 % C alloy using non-polarised neutrons [7].

The nuclear inhomogeneities show a scattering (at $H = 2.5$ T, Fig. 4, curves 2 and 3) as a fractal surface ($\alpha = 3.4\pm0.2$) for large length scale $r > 100$ Å with some mass fractal substructure in the length scale of 10–100 Å ($\alpha = 2.0\pm0.1$). For large q, the 2D scattering patterns obtained for the Fe — 30.5 % Ni — 1.5 % C (wt. %) alloy at $H = 0$ and $H = 2.5$ T show that the magnetic field causes an anisotropy of 2D scattering, thus indicating some magnetic contribution from small length-scale inhomogeneities (Fig. 3) which cannot be destroyed at all even in $H = 2.5$ T.

Thus, we have shown the contribution from magnetic inhomogeneities of different

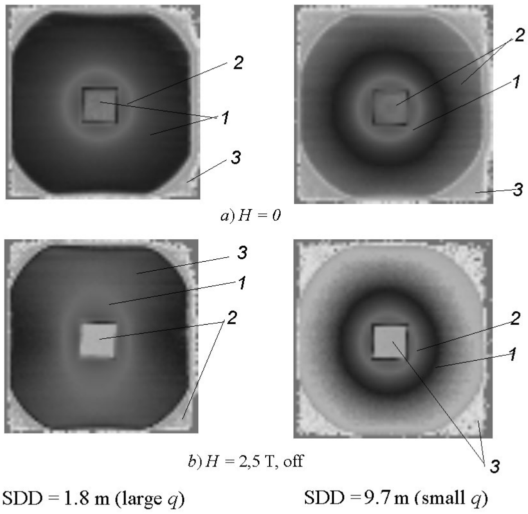

Fig. 3. Examples of 2D scattering patterns in log scale for the Fe — 30.5 % Ni — 1.5 % C (wt. %) alloys in the case of polarized neutrons obtained at sample-to-detector distance of 1.8 m (large q) and 9.7 m (small q): (a) $H = 0$ and (b) $H = 2.5$ T. The curves *1* mean high intensity and the *2* and *3*, lower ones.

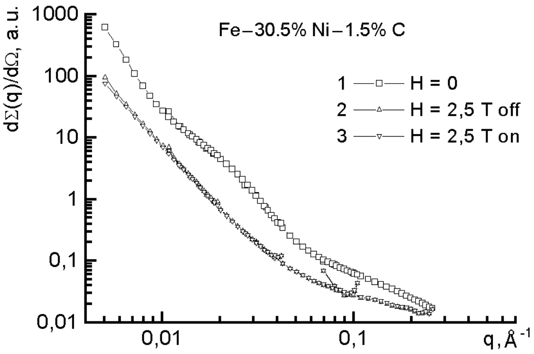

Fig. 4. Polarized neutrons SAS (averaged by φ-angle and summarized by spin direction) in the Fe — 30.5 % Ni — 1.5 % C (wt. %) alloy (for $H = 0$ and $H = 2.5$ T) after annealing of sample at 1100°C in vacuum for 30 min.

sizes and structure to SANS in the Fe–Ni and Fe–Ni–C alloys. We assume that the magnetic inhomogeneities result from fluctuations of the short-range order of interstitial and substituting atoms and mixed-exchange interaction between atomic pairs ($J^{(2)}_{\text{Fe–Fe}} < 0$, $J^{(2)}_{\text{Fe–Ni}} > 0$, $J^{(2)}_{\text{Ni}} > 0$ [16]). Carbon intensifies this trend. The obtained results are consistent with the Mössbauer data indicating the distribution of the hyperfine magnetic fields in the Fe–Ni–C alloys in wide range (5–30 T, [5–7]). The magnetic SANS at $q > 0.1$ was stated in the Fe — 30 % Ni and Fe — 32 % Ni alloys non-enriched and enriched with ^{54}Fe, ^{58}Ni, and ^{62}Ni isotopes in order to enhance contrast and the estimated dimension of spin fluctuations was approximately 10–12 Å [3] that is consistent with our evaluations (20 Å) for close value of scattering vector (Table).

To conclude, the magnetic structure of the f.c.c. Invar-type Fe — 30.3 % Ni and Fe — 30.5 % Ni — 1.5 % C alloys has been analyzed using the small-angle scattering of polarized neutrons. This structure is very sensitive to the applied magnetic field of 2.5 T that results in the reduction of average scattering intensities, changing the slope of the SANS curves, and anisotropy of 2D scattering pattern. SANS in studied alloys annealed at 1100°C results from both chemical fluctuations and magnetic inhomo-

Table. The composition of the alloys and the power law exponent α

Alloy, [wt. %]	H, T	q range [Å^{-1}]	α
Fe-30.3 % Ni	0	0.006–0.01	2.3±0.1
		0.01–0.03	1.6±0.1
		0.05–0.3	1.9±0.1
	2.5	0.006–0.02	3.2±0.1
		0.1–0.3	0.5±0.1
Fe-30.5 % Ni-1.5 % C	0	0.006–0.01	4.1±0.2
		0.01–0.02	2.6±0.1
		0.02–0.06	3.3±0.1
	2.5	0.006–0.03	3.4±0.2
		0.03–0.3	2.0±0.1

geneities. The magnetic inhomogeneities of 16 to 200 Å size (Fe — 30.3 % Ni) and 20 to 300 Å size (Fe — 30.5 % Ni — 1.5 % C) are described in terms of fractal-like structures. The predominant small length-scale magnetic inhomogeneities in the alloys react on the applied magnetic field. Further experiments with Invar Fe–Ni–C alloys at low temperatures are necessary.

Acknowledgments. This study was carried out under support of GKSS Research Centre and the Science and Technology Center in Ukraine (Project #2412).

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Малокутове розсіяння нейтронів в інварних Fe–Ni–C сплавах у магнітному полі

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Досліджено малокутове розсіяння нейтронів (МКРН) в інварних Fe–Ni та Fe–Ni–C сплавах з застосуванням поляризованих нейтронів. Вимірювання проведено без магнітного поля та з прикладеним до зразка магнітним полем 2,5 Т перпендикулярним до пучка нейтронів. Отримано нелінійну криву МКРН в інтервалі векторів розсіяння q = 0.006–0.025 Å^{-1} і проаналізовано з використанням степеневої функції. Продемонстровано суттєвий магнітний внесок у розсіяння нейтронів. З використанням непрямого фур'є-перетворення, оцінено розмір неоднорідностей.