DESY Summer Student Project

Bc. Jana Gamcová
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Date of birth:
August 3, 1986

Nationality:
Slovak

Thermal stability of amorphous microwires studied by in-situ high-energy X-ray diffraction

Area of research:
Photon Physics, Material Science

Time frame:
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Supervisor:

Dr. Jozef Bednarcik
Deutsches Elektronen Synchrotron DESY
Notkestr. 85
22607 Hamburg, Germany
email: jozef.bednarcik@desy.de
phone: +49-40-89981861

Short description of the research project

The aim of this project was to characterize the structure of as-prepared metallic glass based on Fe, Ni, Si, B and follow evolution of its structure during thermal loading using in-situ hard X-ray diffraction. Also influence of alloy composition was studied. Experiments were performed at the BW5 beamline.

Contents

1 Introduction and scientific background

Glass-coated amorphous microwires have attracted considerable attention along the last years owing to their outstanding magnetic properties that make them suitable to be used as sensing elements in sensor devices. Microwires are fabricated by quenching and drawing Taylor-Ulitovski technique, that allows us to obtain tiny ferromagnetic metallic wires (1-30μm in diameter) covered by insulating Pyrex glass coating (2-15 μm in thickness). In particular, positive magnetostriction amorphous microwires exhibit bistable magnetic behaviour which is characterized by a square shaped hysteresis loop and their magnetic properties are very suitable to be employed as sensing elements in devices for technological applications [1]. It is shown that the magnetization process in the presented samples runs mainly through the domain wall motion and it induces interesting magnetic behaviour, which was studied in my bachelor work [2].

2 Goals of the research project

The purpose of X-ray power diffraction (XRD) experiments was to investigate atomic structure of glass-coated $Fe_{77.5-x}Ni_xSi_{7.5}B_{15}$ (x = 75.5, 38.8, 27.9 at %) microwires in as-prepared state and characterize the structure of amorphous samples during thermal loading.

Main goals of the proposed project can be formulated as follow:

- Investigate atomic structure of the set of samples.
- Make the phase analysis.
- Calculate the grain size using Scherrer formula.

3 Experimental part

3.1 Preparation of samples

Microwires are manufactured by means of a modified Taylor-Ulitovsky process based on direct casting from the melt. In the laboratory process, a few grams of the master alloy with the desired composition is put into a Pyrex-like glass tube and placed within a high-frequency inductor heater. The alloy is heated up to its melting point. While the metal melts, the portion of the glass tube adjacent to the melting metal. A glass capillary is then drawn from the softened glass portion and wound on a rotating coil. At suitable drawing conditions, the molten metal fills the glass capillary and a microwire is thus formed where the metal core is completely coated by a glass shell [3].

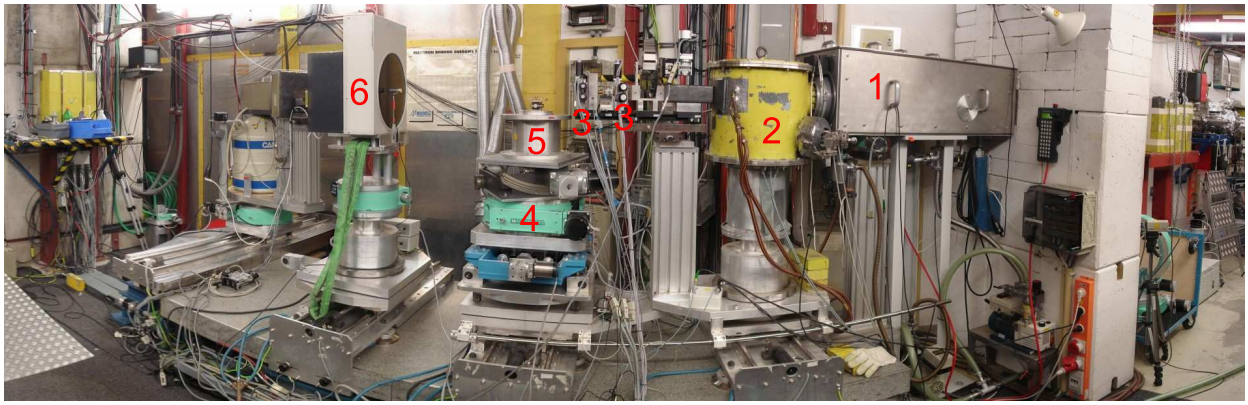

Figure 1: Beamline BW5: 1 - collimator, 2 - monochromator, 3 - slits, 4 - goniometer , 5 - place for sample , 6 - image-plate detector.

3.2 In-situ X-ray diffraction experiments

The measurements were carried out with the multipurpose diffraction instrument at the high-field wiggler, high-energy beamline BW5 at HASYLAB (fig.1). Beamline BW5 is dedicated to X-ray scattering experiments at photon energies between 60 and 150 keV. The large penetration depth at these energies of typically several mm to cm allows the investigation of bulk materials and complex sample environments. This beamline is equipped with an on-line image-plate area detector (Mar345 image plate) for determination of diffracted photons.

In our experiments samples were measured in quartz capillaries with 2mm in diameter and illuminated during 30 seconds by 1x1mm^2 primary X-ray beam of photon wavelength 0.124Å. During in-situ measurement the amorphous alloy was heated by a mirror infrared furnace starting from the room temperature up to 600^{o}C with the heating rate 5^{o}C/min. In picturer 2 there is a detailed view showing infrared lamp furnace. Black tube sitting between two lamps supports capillary with the sample (fig.2).

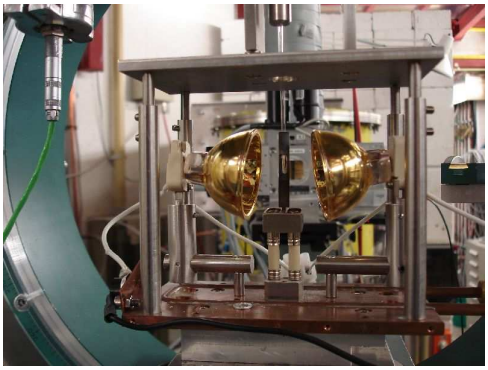

Figure 2: Detailed view showing infrared lamp furnace.

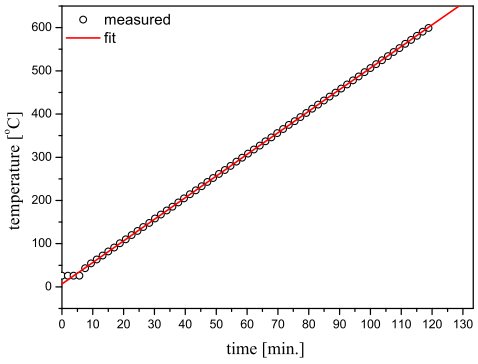

Figure 3: Time dependence of temperature during measuring.

Over thermal loading 63 diffraction patterns were collected at selected temperatures by detector and information from those XRD patterns provided a direct evidence of outgoing structural changes. Sample temperature was controlled by a thermocouple placed inside heat condenser. The images taken from of all samples and different stages of annealing were integrated using Fit2D software [4]. Sample detector distance, with respect to the incoming radiation, as well as precise radiation energy were determined by fitting a standard reference LaB_6 sample.

4 Results and Discussion

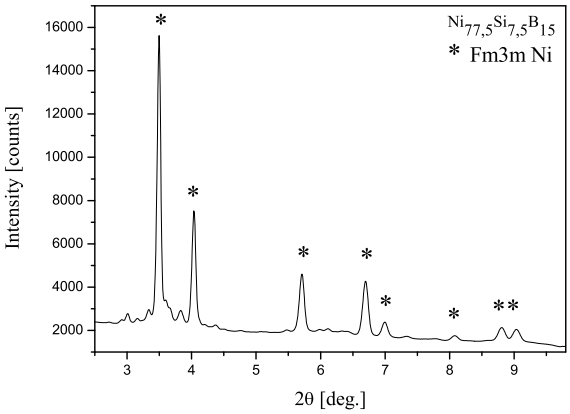

Figure 4: XRD pattern of as-prepared $Ni_{77.5}Si_{7.5}B_{15}$ sample.

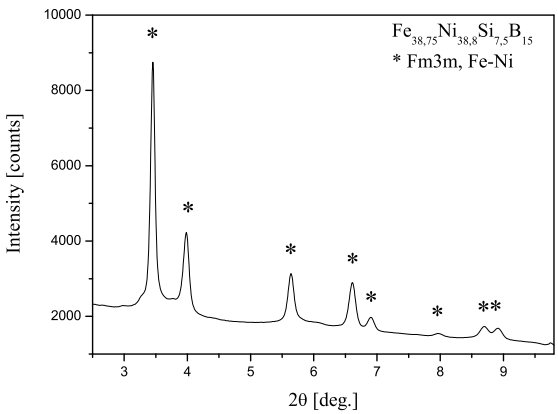

Figure 5: XRD pattern of as-prepared $Fe_{38.75}Ni_{38.8}Si_{7.5}B_{15}$ sample.

Three alloys of microwires, $Ni_{77.5}Si_{7.5}B_{15}$, $Fe_{38.75}Ni_{38.8}Si_{7.5}B_{15}$ and $Fe_{49.6}Ni_{27.9}Si_{7.5}B_{15}$, in as prepared state were measured by X-ray powder diffraction (XRD) experiments at the room temperature. We observed that only one sample is in amorphous state ($Fe_{49.6}Ni_{27.9}Si_{7.5}B_{15}$), the others are nanocrystalline ($Ni_{77.5}Si_{7.5}B_{15}$, $Fe_{38.75}Ni_{38.8}Si_{7.5}B_{15}$) and their diffraction patterns are shown in figure 4 and 5. In nanocrystalline samples phase analysis was done and revealed that Ni - *Fm3m* phase was formed in $Ni_{77.5}Si_{7.5}B_{15}$ alloy and Fe-Ni - *Fm3m* phase in $Fe_{38.75}Ni_{38.8}Si_{7.5}B_{15}$. Both alloys are face center cubic (fig.4).

Grain size D_v

$$D_v = K\lambda/\beta cos(\theta) \tag{1}$$

were calculated using Scherrer formula, where K is Scherrer constant, somewhat arbitrary value that falls in the range 0.87-1.0 (Iassume K=1), λ is wavelength (0.124Å) of the radiation and β is the Full Widht at Half Maximum (FWHM) and it is in radians. Estimated grain size for $Ni_{77.5}Si_{7.5}B_{15}$ and $Fe_{38.75}Ni_{38.8}Si_{7.5}B_{15}$ alloys was 30nm and 11nm, respectively.

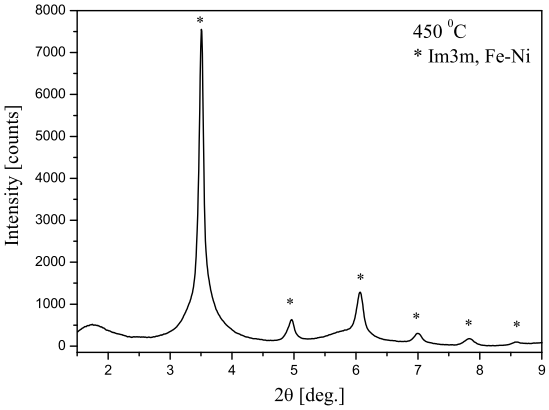

Figure 6: XRD pattern of $Fe_{49.6}Ni_{27.9}Si_{7.5}B_{15}$ sample at temperature 450^0C.

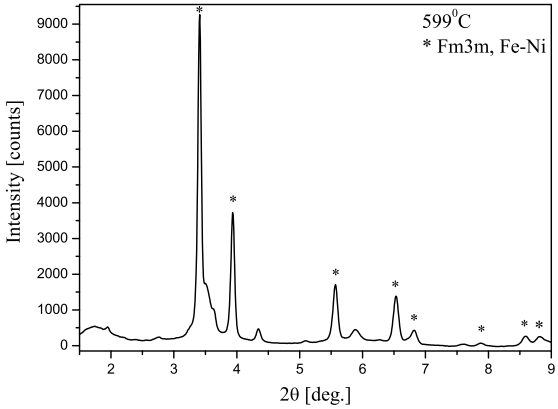

Figure 7: XRD pattern of $Fe_{49.6}Ni_{27.9}Si_{7.5}B_{15}$ sample at temperature 599^0C.

During in-situ measurement the amorphous sample ($Fe_{49.6}Ni_{27.9}Si_{7.5}B_{15}$) was heated by the infrared heater starting from the room temperature up to 600^0C. Figure 12 shows series of diffraction patterns in 3D space acquired during thermal loading. From the room temperature up to 413^0C this alloy is in amorphous state as it can be seen from diffraction patterns, which have only one diffuse maximum. This behaviour is typical for amorphous materials. At the first glance the patterns look the same however careful analysis reveals slight differences. Yavari [5] *at all.* showed that tracing the position of the principal diffraction peaks with the temperature may be used for quantitative analysis of the volume changes:

$$\left\{\frac{q_{max}(T_o)}{q_{max}(T)}\right\}^3 = \left\{\frac{V(T)}{V(T_o)}\right\} = \{1 + \alpha_{th}(T - T_o)\}, \tag{2}$$

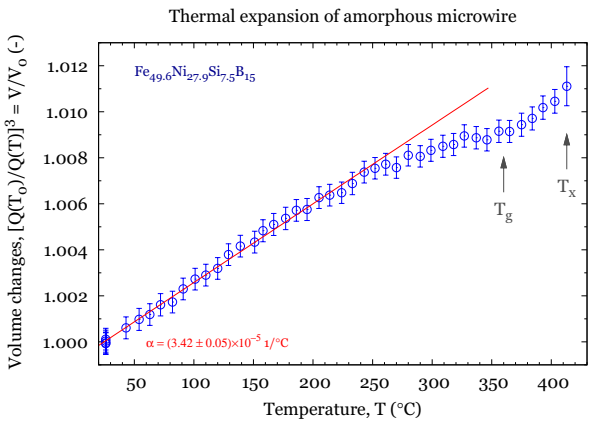

Figure 8: Relative volume changes of $Fe_{49.6}Ni_{27.9}Si_{7.5}B_{15}$ sample in amorphous state.

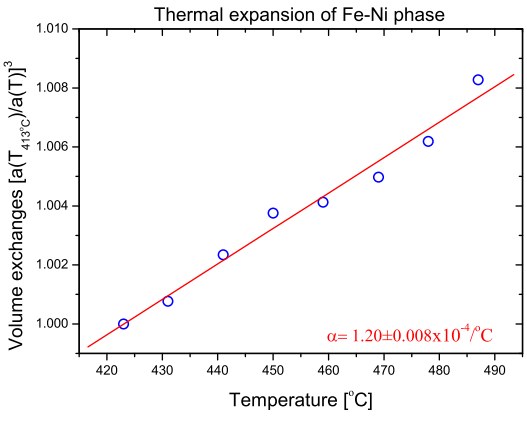

Figure 9: Relative volume changes of $Fe_{49.6}Ni_{27.9}Si_{7.5}B_{15}$ sample in crystalline state.

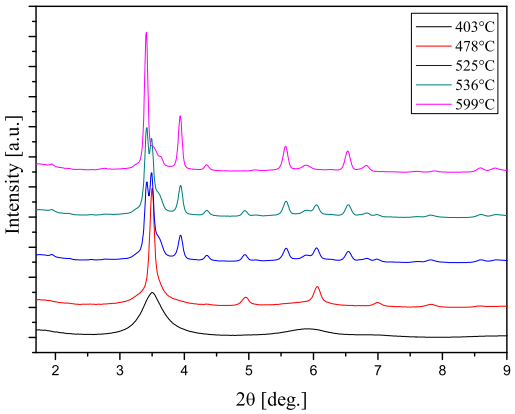

Figure 10: Selected XRD patterns of $Fe_{49.6}Ni_{27.9}Si_{7.5}B_{15}$ alloy. At temperature 478 ^{0}C state of first crystallization, 525 and 536 ^{0}C second crystallization states and 599^{0}C final state.

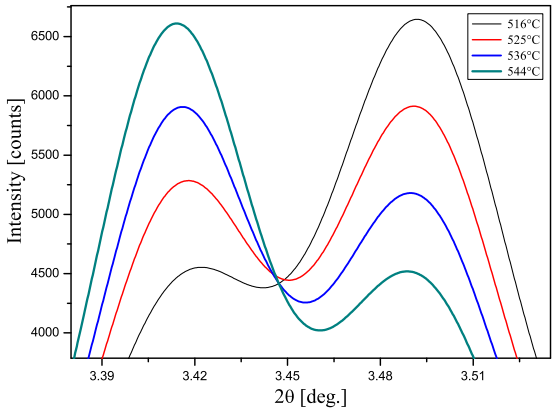

Figure 11: Documentation of phase transformation from Im3m (Fe-Ni) to Fm3m (Fe-Ni) phase in $Fe_{49.6}Ni_{27.9}Si_{7.5}B_{15}$ alloy.

where α_{ht} is the volume coefficient of thermal expansion below T_g as long as no structural change occurs and corresponds to the temperature slope or derivative of the reduced mean atomic volume $\{V(T)/V(T_o)\}$ at T, with the reference T_o corresponding to room temperature.

The profile of the principal diffraction peak was fitted by Pseudo-Voigt function. Figure 8 shows monotonically increase of the sample volume with increasing of tem-

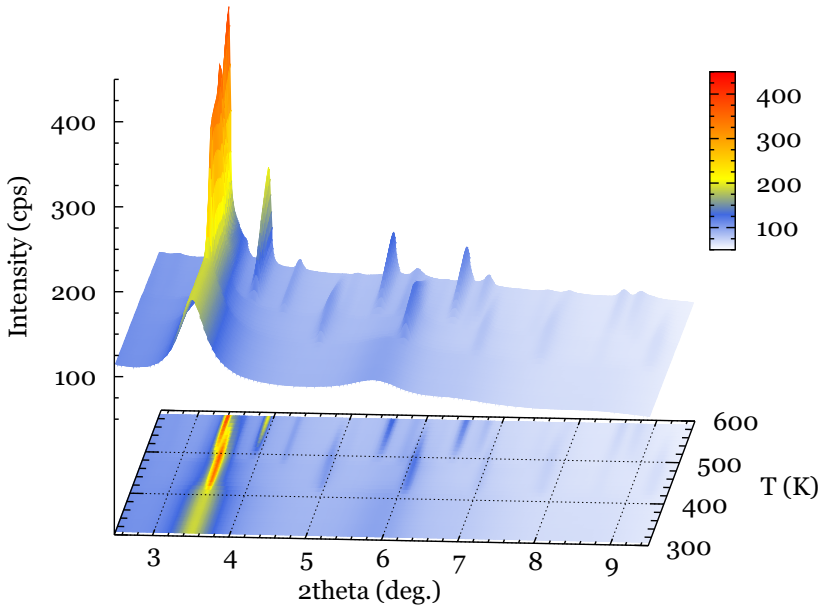

Figure 12: Series of XRD patterns in 3D space for $Fe_{49.6}Ni_{27.9}Si_{7.5}B_{15}$ alloy.

perature up to 260^0C and thermal expansion coefficient was calculated from the slope of volume changes. Its value is $\alpha_{th} = 3.42x10^{-5}\ \ 1/^oC$. Small minimum around 360^0C suggests the presence of the glass temperature T_g. For the comparison in figure 9 the thermal expansion coefficient of crystalline phase, appearing during thermal loading, is shown. Its value is $\alpha_{th} = 1.2x10^{-4}\ \ 1/^oC$.

Additional thermal loading caused significant changes in the structure and first Bragg's peaks were observed at temperature (413^0C) (fig.12), what means that the first crystallization occurred. Those Bragg's peaks were identified as Fe-Ni - *Im3m* phase. Selected diffraction patterns are illustrated in figure 10. At temperature 478^0C Fe-Ni - *Im3m* phase started to vanish and new Bragg's peaks appeared. After careful phase analysis it can be concluded that body center cubic phase Fe-Ni was transformed into face center cubic phase. In more detail between temperatures 516^0C and 544^0C this phase transformation is shown in figure 11.

5 Conclusions

By X-ray powder diffraction (XRD) experiments the atomic structure of measured samples was detected. We observed that one sample is in amorphous state $Fe_{49.6}Ni_{27.9}Si_{7.5}B_{15}$, while the others are nanocrystalline $Ni_{77.5}Si_{7.5}B_{15}$, $Fe_{38.75}Ni_{38.8}Si_{7.5}B_{15}$. We detected in $Ni_{77.5}Si_{7.5}B_{15}$ Ni - *Fm3m* phase with grain size about 30nm in $Fe_{38.75}Ni_{38.8}Si_{7.5}B_{15}$ Fe-Ni - *Fm3m* phase with grain size about 11nm. In the amorphous sample ($Fe_{49.6}Ni_{27.9}Si_{7.5}B_{15}$) during in-situ measurement the phase transformation occurred between body center cubic and face center cubic Fe-Ni phase. The grain size in this alloy is 10nm.

Acknowledgement

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References

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