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Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office using the **Electronic Report Submission Application:**

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Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
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Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** Diffuse Scattering Holography of Ga1-xMnxAs layers	**Experiment number:** HS 3278
Beamline: ID11	**Date of experiment:** from: 11-APR-2007 to: 17-APR-2007	**Date of report:** 11.02.2008
Shifts: 18	**Local contact(s):** Jonathan Wright	*Received at ESRF:*

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Report: Ternary $Ga_{1-x}Mn_xAs$ mixtures possess a high potential in spin electronics because of their ferromagnetic properties incorporated into a semiconductor material (Ohno, 1998). However, at the present state of the growing techniques, no sample with a Curie temperature T_C higher than 170 K can be prepared (Edmonds *et al.*, 2004), while prospective practical applications require further increase of T_C.

In these materials the ferromagnetic coupling is mediated by holes induced by Mn ions in substitutional positions that provides a local magnetic moment and acts as an acceptor. The Curie temperature increases with the concentration of the substitutional Mn cations, but the holes are strongly compensated by defects, the most important being Mn interstitials, which are double donors (Máca & Mašek, 2002). GaMnAs layers have to be grown at low substrate temperatures in order to achieve above equilibrium concentrations of Mn, and low-temperature growth results in high concentration of defects in all GaMnAs samples passivating a large number of holes. Suppression of defects is thus a crucial point for improvement of magnetic properties of GaMnAs layers.

Annealing at temperatures close to the growth temperature increases the Curie temperature as well as the saturation magnetization (Hayashi *et al.*, 2001). Consequently, channeling Rutherford backscattering and particle induced x-ray emission experiments indicated decreased concentration of Mn interstitials after annealing (Yu *et al.*, 2002). The concentrations of substitutional and interstitial Mn atoms can be also estimated by indirect methods, e.g. by measuring lattice parameter (Mašek *et al.*, 2003) or the integral intensities of weak Bragg reflections (Glas *et al.*, 2004; Frymark *et al.*, 2005).

Recently, we studied the GaMnAs layer with low concentration of impurities ($x = 0.02$) by means of the x-ray diffuse scattering holography (Kopecký, 2004; Kopecký *et al.*, 2006), reconstructing the local atomic structure around the impurities and found manganese atoms only in the substitutional positions.

In the experiment carried out on the ID11 beamline we used the x-ray diffuse scattering to study the structural changes in the GaMnAs layer induced by annealing. The sample was mounted on a heating stage with the incident beam almost parallel to the crystallographic axis (110). After collecting the diffuse scattering patterns on the as-grown GaMnAs layer (Fig. 1), the heater was switched on at 200°C for a period of 2 hours, and a second set of diffuse scattering patterns was collected on the annealed layer after cooling the sample down to the room temperature. The real-space images were reconstructed using the Helmholtz-Kirchhoff integral theorem providing the local Patterson function (Kopecký *et al.*, 2003). The reconstruction of the as-grown sample shown in Fig. 2(left) exhibits peaks corresponding to interatomic vectors of gallium arsenide. There is no evidence of interstitial atoms in Fig. 2(left). In other words, possible peaks caused by atoms in interstitial positions are masked by artefacts inherent to the Helmholtz-Kirchhoff reconstruction algorithm (Barton, 1991; Marchesini & Fadley, 2003). However, the same artefacts are present in the real-space reconstruction of the annealed sample, providing almost the same image as the as-grown sample, and can be subtracted. Fig. 2(right) shows the difference of the Patterson functions of the GaMnAs layer before and after annealing. Evidently, the Patterson function of the as-grown layer exhibits peaks corresponding to atoms in interstitial positions that disappear (or decrease) after annealing. Providing Mn are these interstitial atoms, the peaks in the difference Patterson function correspond to Mn-Ga and Mn-As interatomic vectors (Giacovazzo *et al.*, 2002) yielding thus the local neighborhood of Mn interstitials. According to the positions of the nearest neighbours, the interstitial atoms are located in the centre of gallium and/or arsenic tetrahedron as expected (Máca & Mašek, 2002). Although this method is not able to distinguish between these two types of the interstitials since both of them give the same difference Patterson function, the presence of interstitial atoms in as-grown GaMnAs layer, it demonstrated that the concentration of interstitials decreases when the sample is submitted to the low-temperature annealing. This is in accordance with previous studies (Yu et al., 2002; Edmonds et al., 2004) where the increase of TC after annealing is assigned to the out-diffusion of the highly mobile Mn interstitial towards the surface followed by their passivation.

We also carried out in-situ measurements of the behavior of interstitials during the annealing process. However, this part of the experiment has not been successful yet, mainly because the differences in measured diffuse scattering patterns induced by system geometrical instability at higher temperatures exceeded those originated in the structural changes of the GaMnAs layers. An improved sample heater has to be constructed before using this method to the systematic study of GaMnAs layers.

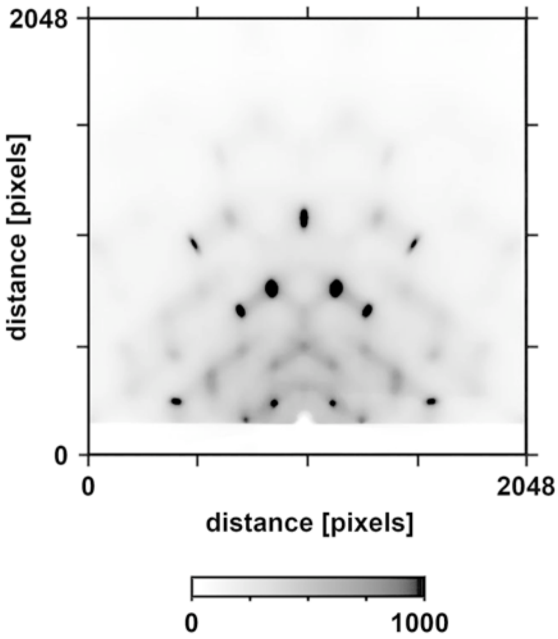

Figure 1 The diffuse scattering pattern of the GaMnAs layer recorded on a CCD camera at a condition of total reflection using the photon energy of 28.3 keV. The bottom part of the image is shielded by the sample. The intensity scale is in counts per pixel of the detector.

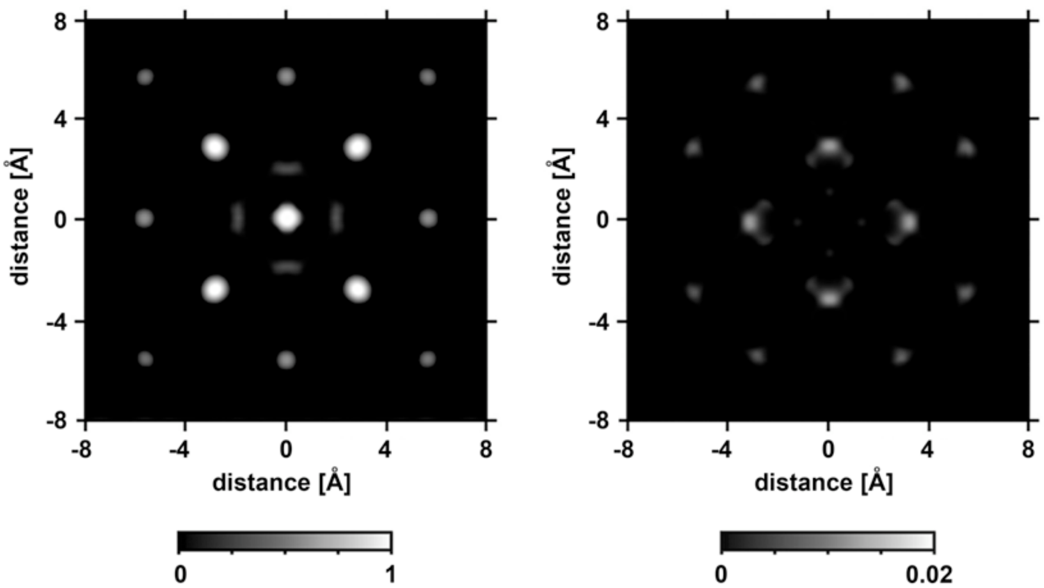

Figure 2 (Left) The real-space reconstruction of the diffuse scattering pattern in Fig. 1 in the plane parallel to the (001) crystallographic plane at z = a. The positions of peaks correspond to the interatomic vectors in GaAs single crystal. (Right) The local neighbourhood of interstitial atoms obtained as the difference of the Patterson functions of the GaMnAs layer before and after annealing.

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