ESRF	Experiment title: Magnetic properties of GeMn quantum dots grown on Si(001)	Experiment number: MA-2708
Beamline:	Date of experiment:	Date of report:
ID32	from: 03/09/15 to: 04/09/15	10/09/15
Shifts:	Local contact(s):	Received at ESRF:
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Objective and expected results:

Strain-induced quantum dots (QDs) may represent a solution to form dilute ferromagnetic semiconductors (DMS) with high Curie temperature (T_C) thanks to the spatial confinement of magnetic atoms. Recently, we have have demonstrated that Manganese-doped Germanium QDs grown at low temperature (<450°C) in order to reduce inter-diffusion processes and containing up to 3% of Mn are exempt of Mn rich phases. SQUID measurements on these samples suggest that T_C is well above room temperature (T_R). The aim of the proposal was to investigate by X-ray Magnetic Circular Dichroism (XMCD) the magnetic properties of these Mn-doped Ge QDs grown on Si(001). The expected results, as stated in the proposal, were to confirm the existence of ferromagnetism in Mn-doped QDs at high temperature and to investigate its origin.

Results and the conclusions of the study:

XMCD experiments have been carried out on the ID32 beamline at ESRF by detecting XAS (x-ray absorption spectroscopy) spectra at the Mn $L_{2,3}$ edges using left and right circularly polarized light in the total electron yield mode. Magnetic fields of up to 1 T were used to magnetize the samples up to saturation. Since only three shifts have been allowed to this project for test measurement, we have focused on one Mn concentration and have investigated the magnetic properties of GeMn QDs containing about 2% Mn. The sample was capped with 1nm of Ge deposited at room temperature in order to prevent QDs oxidation. Due to technical problems with the cryostat cooling, all measurements have been performed above 280K.

Figure 1 shows the XAS spectrum measured at 280K on QDs containing around 2% of Mn. Two absorption regions corresponding to $2p_{3/2}$ (~640 eV) and $2p_{1/2}$ (~650 eV) transitions are clearly visible. Well-defined shoulders appear at 642 and 644 eV and a doublet structure is observed in the $2p_{1/2}$ excitation region. This well-structured spectrum is clearly different from spectra steming from metallic Mn₅Ge₃ phase [1, 2] or from the presence of Mn dimers as found in Ga_{1-x}Mn_xAs alloys [3]. On the other hand, this spectrum is in close agreement with the calculation based on the atomic multiplet theory for Mn²⁺ (3*d*₅) presented by the red dashed line in Figure 1a, which suggests that Mn dilution has been achieved in our QDs. This is also in agreement with accurate first-principles calculations [4]. Such a structure has been observed in diluted Mn impurities in the Ge matrix in an ion implanted Mn–Ge(100) alloy [5]. The XAS multiplet structure cannot be ascribed to oxygen contamination as this would induce a manganese oxide related multiplet structure as observed in reference [6]. This demonstrates that the capping layer was thick enough to prevent the sample oxidation.

The XMCD spectrum (Fig.1b) reveals a negative signal (approximately 640 eV) as well as two small positive signals (approximately 642 eV and 644 eV) in the $2p_{3/2}$ region and also two other positive signals (approximately 651 and 652.5 eV) in the $2p_{1/2}$ region. Since the XAS measurements rule out the presence of metallic Mn phase (such as Mn₅Ge₃ for examples) or Mn oxides, the dichroic signal is attributed to the presence of a DMS phase at high temperature, which is consistent with SQUID-VSM measurements shown in Figure 1c. Furthermore, no manganese oxide (with divalent Mn²⁺) is expected to give such a dichroic signal at room temperature and in 1T. It has also to be underlined that DMS thin films with intrinsic ferromagnetism exhibit Curie temperatures lower than 150K.



<u>Figure 1:</u> a) XAS and b) XMCD spectra measured at the Mn 2p-3d absorption edge of the GeMn QDs layer at T=280 K (black lines); red dotted lines correspond to the-calculations based on the atomic multiplet theory for Mn^{2+} in the 3d₅ configuration; c) Hysteresis loops measured at 10K and 300K with the magnetic field applied in the sample plane. Inset: temperature dependence of the magnetic moment measured in 1T.

These first XMCD experiments on GeMn QDs have brought interesting preliminary results. We have demonstrated with soft Mn $L_{2,3}$ XAS that the electronic configuration of the Mn impurities in Ge is Mn²⁺ (3*d*₅). The presence of dichroism at room temperature demonstrates that the spatial confinement of Mn atoms induces a ferromagnetic order above room temperature. To our knowledge, it is the first time that the relationship between high Curie temperature (above T_R) and dilute magnetic phase is unambiguously evidenced. New shifts using XMCD are highly needed in order to continue the study of this system and to compare the properties of DMS QDs and thin films for various Mn concentrations.

Justification and comments about the use of beam time:

Both the extreme sensitivity and the element specificity of XMCD are highly needed to study the magnetic properties of our system. Low temperature is necessary to improve the signal-to-noise ratio of the dichroic signal and also to measure the magnetic properties of DMS thin films with comparable Mn content for which the Curie temperature stand below 150K. Unfortunately, due to technical problems, the temperature in the cryostat could not go down below 280K, which is insufficient to reach the aims of this proposal. For instance, an acquisition time of 10 hours was required to register a XMCD spectrum with such a low dichroic signal.

References

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