Report of the experiment HE-2439

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(Dated: February 28, 2008)

In the present report, we will describe the main goals and results of HE-2439 experiment that was conducted in September 2007 on the beamline ID24 at ESRF. We will briefly describe the underlying scientific question that we wanted to answer with the proposed experiment and we will join a detailed resume of the experimental objectives that have been completed during the beamtime together with a list of unsuccessful experimental trials. If many unexplored aspects of the transition metal silicides system have been clarified with the data collected during the experimental period, the main objective could not be totally completed due to major technical issues connected with the experimental setup. Considering the importance of the scientific question that could be answered completing the proposed experiment and the significant recent technical improvement in ID24 XMCD setup, we hope to be able to have an extra beam time in the near future to conclude our work.

I. INTRODUCTION

The central scientific issue that convinced us to apply for a beam time at ESRF is the still unclear origin of the magnetic structure in transition metal monosilicides. The phase diagram of this family is quite rich and the magnetic behavior of various members changes drastically upon doping and depending by the transition metal (TM) element. In particular, the impact of Coulomb interactions in the d-shell of the TM atom on the magnetic behavior of these compounds remains unclear. Among this silicides family there are two compounds which present similar low temperature behavior: MnSi and $Fe_{0.6}Co_{0.4}Si$. In both cases, the low temperature state is characterized by an helimagnetic structure which has been demonstrated, using neutrons and transport experimental techniques, to vanish upon application of hydrostatic pressure. In this regarding, the facilities available on ID24 seemed to be the best tools to study the evolution of such transition: there in fact it is possible to acquire spectral information as a function of pressure measuring the XANES signal at low temperature.

In order to detect the magnetic signal a dichroic measurement (XMCD) was necessary. It is clear that already at this stage, the experiment appears quite challenging because it requires the combination of various techniques with extreme precision and stability requirements. As a further complication we want to point out here that the magnetic information that could be extracted from such an experiment would only be an indirect information due to the hybridization between p and d orbitals in TM ion. Since a measurement at the K-edge is mostly sensitive to transitions into p-states we expect the XMCD signal originated by processes in the d-band, to be extremely small.

Assuming that such a small signal could be detected, following its evolution under pressure and combining these results with the pressure dependence of the electronic structure (cluster LMTO band calculations) should allow us to give a solid contribution to the debate about the origin of the magnetic phase in transition metal mono silicides.

II. DISCUSSION OF EXPERIMENTAL RESULTS

Before discussing the XMCD measurement itself we would like to underline here the fact that the literature lacks of any experimental data regarding standard x-rays measurements on TMSi; for this reason part of the time was then dedicated to measure the temperature and pressure dependence of the XANES signal. Anyhow these kind of measurement were planned as being "preparatory" experiments for the main one. The original plan was to measure MnSi first and then $Fe_{0.6}Co_{0.4}Si$ .Being a pure binary system, MnSi was considered cleaner and thus of more fundamental interest.

The pressure measurement on MnSi had to be done using special drilled diamond culets in order to avoid excessive absorption at the Mn K-edge energy. During this part of the experiment we were able to measure the temperature dependence of the XANES signal and its low temperature pressure dependence. Unfortunately right before the beginning of the XMCD experiment one of the two diamond broke and it was not possible to continue.

The analysis of the collected data gave interesting results which are partially shown in Fig. 1: in particular it was possible to calculate using FEFF8.4 the expected XANES signal at the K-edge and we found extremely good agreement with the experimental data[?] . Somehow this result was quite unexpected since the same calculation on the L-edge did not gave the same good agreement unveiling the possible strong correlation among d electrons this system. Such a correlation does not seem to be strongly mirrored on the p-electron states even in presence of hybridization. The following set of measurements done on $Fe_{0.6}Co_{0.4}Si$ was certainly more successful. In fact we were able to extract the complete high and low temperature pressure dependence of the XANES signal

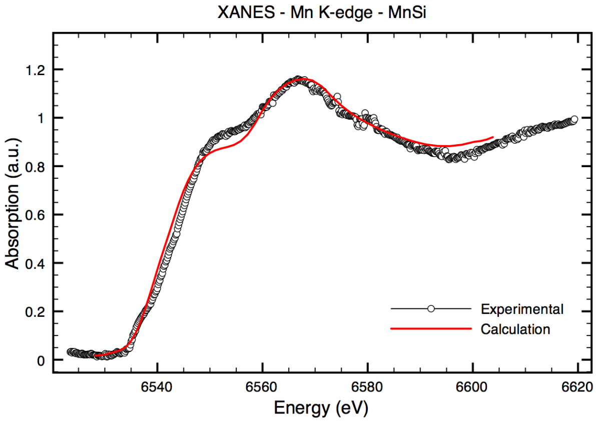

FIG. 1: Comparison between the experimental XANES data for Mn K-edge (black open circles) and theoretical calculation done with FEFF8.4 (red line).

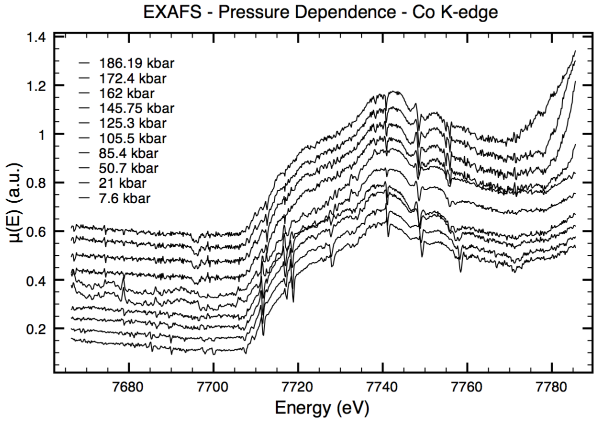

FIG. 3: Pressure dependence of the XANES absorption spectra for Co K-edge in $Fe_{0.6}Co_{0.4}Si$ up to 18.6 GPa. The quality of the data is strongly affected by the degradation of the silicon plate used as monochromator.

to pressure up to 20 GPa both for the Fe K-edge and for the Fe K-edge (Fig. 2 and Fig. 3). Here the experiment was technically easier because of the higher photon flux at the Fe and Co edges but nevertheless extremely time consuming due to the need of a complete realignment of the system in order change between Fe and Co absorption edge energy.

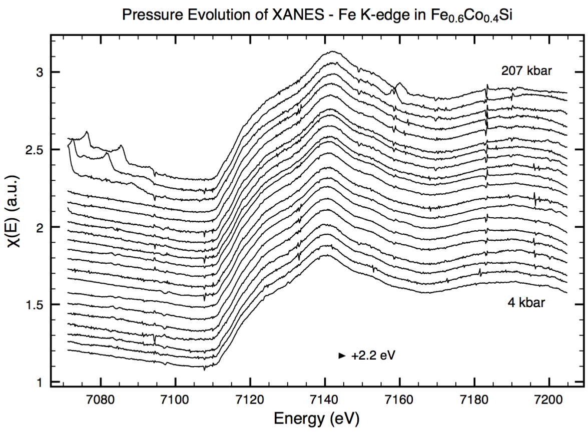

FIG. 2: Pressure dependence of the XANES absorption spectra for Fe K-edge in $Fe_{0.6}Co_{0.4}Si$ up to 20.7 GPa.

Concerning the XMCD measurements on this compound, which we estimated to be the most difficult (but also the more interesting) part of the experiment, we tried to do the measurement on both the Fe and the Co edges with contrasting results.

The main idea of the experiment was first to cool down the sample in the pressure cell in order to bring it in the helimagnetic state, then to take two measurements with circularly polarized light for two magnetic field orientations. The XMCD signal should then appear as the difference between the two absorption spectra. Repeating the measurements at various pressure should allow us to track the evolution of the magnetic state in the system and to extract consequently important physical informations.

From the very beginning the signal appeared to be quite unstable but in the case of Fe edge in $Fe_{0.6}Co_{0.4}Si$ we were able to detect it although very low as expected ($\sim 4.1 \times 10^{-4}$). A comparison between the XMCD signal measured inside the pressure cell during our experiment and a signal obtained in a standard XMCD setup (ID12) is presented Fig. 4. This graph demonstrates that despite the complex setup and the low signal the successful completion of the proposed experiment was practically possible.

Unfortunately the system started to be more and more unstable due to temperature stability issues probably caused by eddy currents in the heater's coil of the cryostat and to the degradation of the quality of the bent silicon plate used as polychromator. This latter issue was certainly determinant for the failure of the measurement of the XMCD signal on the Co edge. The effect of poychromator degradation on the signal is visible (glitches) in Fig. 3, where the room temperature pressure dependent XANES signal is reported.

If this last measurement could have been successfully completed, a lot of interesting information could have been certainly extracted. Table I summarizes the measurements that were planned during the beam time with a cross indicating their completion.

Despite the unsuccessful result on the XMCD side, a complete EXAFS study of these 2 materials has been carried out[1]. In the associated paper, the structural disorder represented by the mean square relative displacement between spectator atoms was derived from comparison of

Compound	MnSi	$Fe_{0.6}Co_{0.4}Si$	
Edge	Mn	Fe	Co
Room Temperature (RT) EXAFS	X	X	X
Low Temperature (LT) EXAFS	X	X	X
Pressure dependent XANES (RT)	X	X	-
Pressure dependent XANES (LT)	X	X	-
Ambient pressure XMCD	-	X	-
Pressure dependent XMCD (LT)	-	-	-

TABLE I: Measurements performed during HE-2439 experiment.

experimental data with cluster simulations and L and K-edge data. This analysis seems to confirm the key role on the magnetic properties of valence state of the TM d electrons. In particular the Debye-Weller factor temperature dependence between absorbing atoms and the surrounding silicon atoms seems to be correlated with the valence state of the TM atom confirming a net difference between the MnSi and FeSi cases with respect the CoSi Case.

As anticipated the XANES measurement were also used to discuss the valence states of the transition metal ion in transition metal monosilicides completing a broader study involving previous L-edge XAS measurements and valence band photoemission spectroscopy[2].

A. Technical problems

A crucial question emerging when we started the experiment was the possibility to tune the pressure inside the diamond anvil cell (DAC) with sufficient precision: in fact at low temperatures, magnetic transitions in these compounds occur around 1.5 GPa for MnSi and 5.5 GPa for $Fe_{0.6}Co_{0.4}Si$ which are very small pressures if compared with the maximum pressure reachable with the DAC apparatus. We experimentally checked the feasibility of such regulation but due to a strong fluctuation of the temperature in the cryostat the pressure on the sample was probably also varying moving the system below and above the transition pressure. The main reason was that due to the oscillation of the temperature close to the sample, the pressurizing gas present in the DAC capillar was also expanding and contracting thus causing a fluctuation in the applied pressure on the sample.

The XMCD signal at the K-edge is the signature of the hybridization of p orbitals with the d-shell. As seen in Fig.4, the amplitude of this signal is very small (4.10^{-4} in $Fe_{0.6}Co_{0.4}Si$) and the detection is made possible only because of the high performance of the source and of the equipment available at ID24. Here, in addition to small signal, we used a pressure cell, a magnet and a low temperature cryostat which was introducing noise in the signal. Due to the mentioned requisites, long integration times are required in order to increase the signal to noise ratio making the measurement very sensitive to fluctuations of the source. As already mentioned, many technical problems have been encountered during the experiment, mainly due to the fact that all the equipments were often used at their limits.

From the acquired experience we can say that the main goal of the experiment was virtually possible to achieve but due to specific instabilities mostly due to the cryostat and to the monochromator it was not possible to successfully complete the proposed experience. However the positive indications that we collected during the beamtime and the possibility to solve the cited problems in a relatively short time thanks to the contribution of the beamline scientists, convinced us to reapply for an extra beam time.

B. The XMCD signal is there!

Fig.4 shows a comparison of XMCD signal measured in the pressure cell during our beam time and a record done afterwards on ID12.

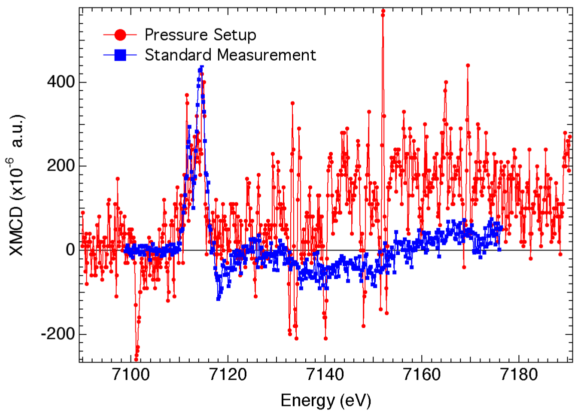

FIG. 4: XMCD signal at Fe-edge in $Fe_{0.6}Co_{0.4}Si$ on ID12 (blue) and in the pressure cell on ID24 (red).

III. REASONS TO CONTINUE

A big effort has been carried out during these 27 shifts of beam time both in the understanding of physical properties of transition metal silicides as well as in the development of the XMCD experimental setup on ID24. In conclusion all the team that worked day and night hoping for a successful conclusion felt really close to the goal but due to the described technical issues was not possible to observe the suppression of the magnetic state with the pressure. Now it seems that many technical questions have been answered making us confident in a possible second run which will be, strong of the acquired experience, most likely the winning one.

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[1] K-edge x-ray absorption spectroscopy on MnSi and $Fe_{0.6}Co_{0.4}Si$, R. Tediosi, J. Teyssier, E. Giannini, R. Viennois, D. van der Marel, O. Mathon, and S. Pascarelli, to be submitted to Physical Review B

[2] The suppression of valence fluctuations in transition metal mono silicides, F. Carbone, R. Tediosi, M. Zangrando, J. Teyssier, E. Giannini, F. Parmigiani, Th. Jarlborg, D. van der Marel, O. Mathon, and S. Pascarelli, to be submitted to Physical Review B