High pressure studies of magnetism

We have been studying during the two last years, Pt based system under pressure by XMCD at ESRF on the ID24 beam-line. The first year we focus on the Pt3Cr system at the Pt L_{23} edges.
Here we recall the main results and arguments of the previous experimental report.
The Pt_3Cr system has been chosen as model system. In fact, recent work[1,2] reports on the magnetism of Pt in this alloy which is very unusual; the L_3 and L_2 XMCD signals have the same sign, meaning a dominant orbital contribution. Pt_3Cr is therefore a good candidate to investigate volume-dependent magnetism of Pt. In addition the Curie temperature is 450 K, well above the room temperature. Moreover, only the crystallographically ordered phase carries a magnetic moment. A great pressure sensitivity of the magnetic moment is therefore expected because changes in crystallographic phase or order can be induced.
Both L_3 and L_2 edges XMCD have been measured under pressure, thereby probing the 5d Pt magnetic moment. X-ray diffraction under pressure doesn't show any change in the crystallographic structure of Pt_3Cr, which remains cubic. The XMCD signals under pressure at the Pt L_3 and L_2 edges (figure1) show a slow decrease between 0 and 3.5 GPa followed by a very abrupt increase between 3.5 and 5.0 GPa.

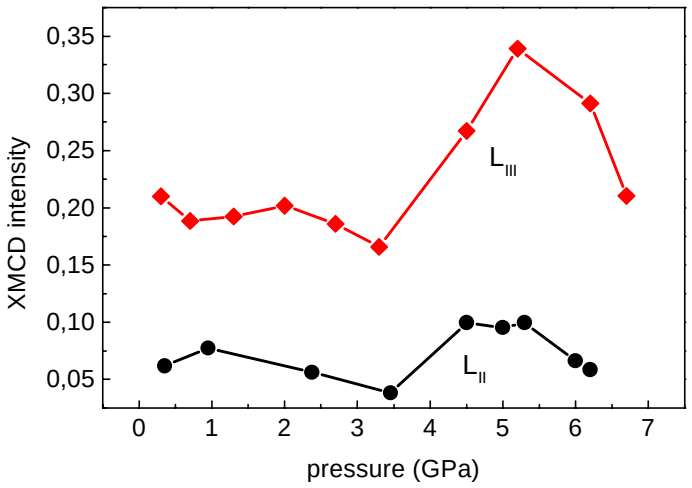


Figure1: Integral of Pt L_3 and L_2 XMCD signals on Pt_3Cr as a function of pressure.

The first decrease reflects an attenuation of the magnetic moment carried by the Pt 5d states along the x-ray propagation axis. It could originate from a decrease of the Cr magnetic moment, inducing a smaller Pt magnetic moment, or from a progressive change of the magnetic order resulting in a smaller ferromagnetic component.
The following increase of the platinum 5d band magnetic moment is induced by the increasing hybridization with the 3d Cr band. This second effect is probably already present between 0 and 3.5 GPa but is hidden by the decrease of the total projected moment. After 5 GPa there is a rapid attenuation of the signal, down to zero which could be due to the loss of the crystallographic order in the sample.
The main result of this study is the constant branching ratio along the pressure domain. There is no L/S ratio variation between 0 and 5 GPa which is rather surprising considering the strong magnetic moment dependence from crystallographic order. The disappearance of the

magnetic moment with the loss of crystallographic order shows the great sensitivity of the orbital and spin moment to the local order. At the same time, their ratio remains insensitive to the reduction of interatomic distance between 0 and 5 GPa (which is typically ~ 0.7%).

The loss of crystallographic order after 5 GPa must be studied in more detail by x-ray diffraction. It could be sample preparation dependent and to be overcome in the next future. This could allow the measurement of XMCD signals at higher pressures up to the orbital moment quenching.

During the last year we take full benefit of our allowed beam time on ID24 thanks to the great recent improvement of the beam line. First we confirm the results of the previous year on Pt_3Cr at both edges. We start the study of other compounds i.e. $Pt_3Cr_{0.5}Mn_{0.5}$ and $Pt_3Cr_{0.75}Mn_{0.25}$. figure2.

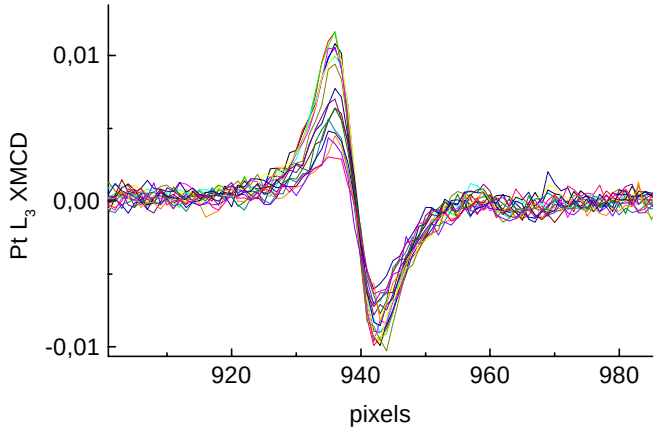
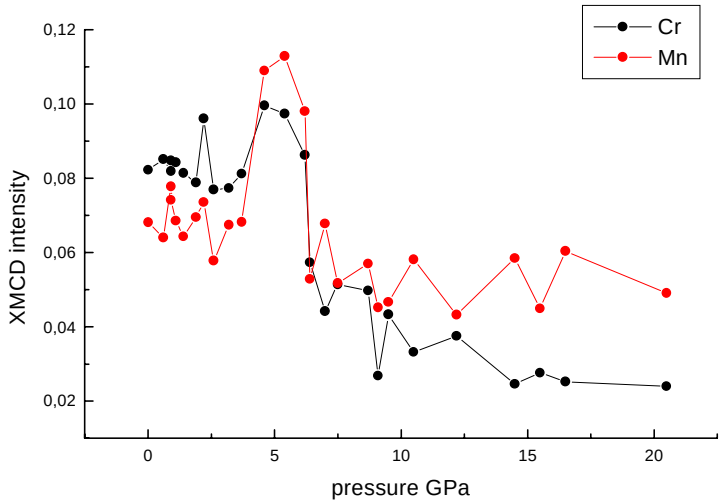


figure2. Pt L_3 XMCD of $Pt_3Cr_{0.5}Mn_{0.5}$ at various pressure

The L_3 XMCD signal of $Pt_3Cr_{0.5}Mn_{0.5}$ is very interesting because we can separate the Cr and Mn contribution to the magnetic polarization of the 5d band of Pt. The positive part is due to Cr and the negative one to Mn. The same append for $Pt_3Cr_{0.75}Mn_{0.25}$ with a ratio proportional to the stoichiometry.
On figure 3 we report the absolute value of the amplitude of the Cr and Mn part of the XMCD signal in $Pt_3Cr_{0.5}Mn_{0.5}$. Both show the same pressure behavior as describe for Pt_3Cr, i.e. an increase following by a large decrease. Nevertheless the signal doesn't go down to zero as for Pt_3Cr. This could be due to the presence of Mn, (the magnetic moment in Pt_3Mn is know to not being sensitive to the crystallographic order), inducing a magnetic moment on both Pt and Cr in the high pressure regime in $Pt_3Cr_{0.5}Mn_{0.5}$.
We observe a larger decrease for the Cr than for the Mn contribution of the Pt signal witch agree with a smaller sensitivity to the crystallographic order of the Mn magnetic moment than the Cr one.

We measure also the pressure variation at the L_2 edge of Pt in $Pt_3Cr_{0.5}Mn_{0.5}$. We obtain of course on positive peak varying with pressure in the same manner as seen in figure1 for Pt_3Cr. A small decrease of the signal between 0 and 3.5 GPa, following by a large increase between 3.5 and 5 Gpa and then a large decrease after 5 GPa. Here again the signal is largely reduced but don't go to zero at high pressure like in for the L_3 edge.



During the last day of the allowed beam time we focus on the main unresolved experimental problem of our study, the decrease of the signal after 5 GPa. We suspect a loose of perfect hydrostaticity after 5GPa giving non-physical decrease of the XMCD signal. We try to change our pressure-transmitting medium from silicon oil to an ethanol/methanol mixture, witch is know to be more hydrostatic than silicon oil above 5 GPa.
Unfortunately our gasket of tantalum opened during the pressure increase just at 5 GPa. The very short remaining beam time prevent to prepare an other sample for an second experiment, so we try again with the same sample and a new gasket with a smaller hole, witch give a better chance to not open. The result vas very surprising, first we see that the first increase of XMCD is fully reversible and reproducible. Secondly we didn't observe any decrease of the XMCD after 5 GPa, but a continuous soft increase up to 20 GPa, were the gasket opened again.
We can't conclude today if the change of XMCD pressure behavior is due to the change of pressure-transmitting medium or to the fact that the sample has already seen a first increase of pressure. We already observe two years ago the same change in the XMCD behavior at the L_3 edge of Pt in Pt_3Cr in a second pressure increase on the same sample.

We hope to complete the large set of reliable results obtained between 0 and 5 GPa by new measurements above 5 GPa with a new pressure method. We propose to focus on the $Pt_3Cr_{0.5}Mn_{0.5}$ at the L_{23} edges. We hope to follow up to 25 GPa the spin and orbital magnetic moment behavior of Pt induced respectively by Mn and Cr. The pressure technique will be based on alcohol-mixture and liquid argon as pressure transmitting medium and rhenium gasket.

References

1. W. Grange et al., J.Sychrotron Radiation 1999
2. Maruyama et al., JMMM 1995, **140-144**, 43-44