EUROPEAN SYNCHROTRON RADIATION FACILITY
ESRF User Office
BP 220, F-38043 GRENOBLE CEDEX, France
Delivery address: 6 rue Jules Horowitz, 38043 GRENOBLE, France
Tel: +33 (0)4 7688 2552; fax: +33 (0)4 7688 2020; email: useroff@esrf.fr; web: http://www.esrf.fr

Experimental report on Proposals HE 3667 ID12 : 29-09 / 05-10-2011

Induced magnetic moment from iron on 5d states in FePt and FeAu clusters assemblies

Scientific background

Bimetallic magnetic nanoparticles (NPs), where the Magnetic Anisotropy Energy (MAE) can be tuned, are excellent candidates for potential biomedical applications or in spintronics. Nevertheless, in view to make further progress in the optimization of NPs and to better understand the origin of unusual magneto-optic response of core/shell FeAu and high MAE in FePt NPs, we performed X ray Magnetic Circular Dichroïsm (XMCD) measurements at the Au and Pt edges on ID12 to determine the induced magnetic moment from iron on 5d states and to compare with XMCD measurements recently performed at the Fe L_3 edge on the same samples. Indeed, we would like to verify experimentally that magnetism can be the driving force for the stability of well defined local-ordered nanoalloys as suggested by ab-initio calculations.

Sample preparation and characterizaton

Our samples are prepared from NPs pre-formed in the gas phase and then codeposited in UHV with the matrix. Adjusting independently the deposition rate of the nanoparticle and matrix beams, very diluted (< 1% of NPs in volume) clusters assembled films, with negligible magnetic interaction among NPs, are obtained. By coupling an electrostatic quadrupole deflector to the setup, we have been able to deposit mass-selected iron based nanoalloys (FeAu and FePt) with relative cluster size dispersion to about 7%**.** We focus our attention to inert amorphous carbon matrix with a weak influence on the interface magnetic anisotropy in order to preserve and investigate the intrinsic cluster properties. The as-prepared carbon coated mixed nanoparticles were previously characterized using high resolution transmission electron microscopy (HRTEM) and x-ray techniques. For all compositions, the nanoparticles presented a mean diameter of about 2 nm and crystallize in the FCC chemically disordered phase. Because post-annealings are compulsory to generate ordered phases and/or segregation (core-shell effects), all the mass-selected samples were studied as-prepared and after annealing up to 550°C.
Prior to experiments on ID12 beam line, XMCD measurements at the Fe $L_{2,3}$ edge have been performed on the same samples at SLS together with complementary SQUID magnetization measurements at Lyon. These experiments revealed a global rise in the blocking temperature and in the spin magnetic moment per Fe atom, for the annealed nanoparticles. The origin of such an enhancement, being still an open question, will be closely related to the induced moments at the 5d sites.

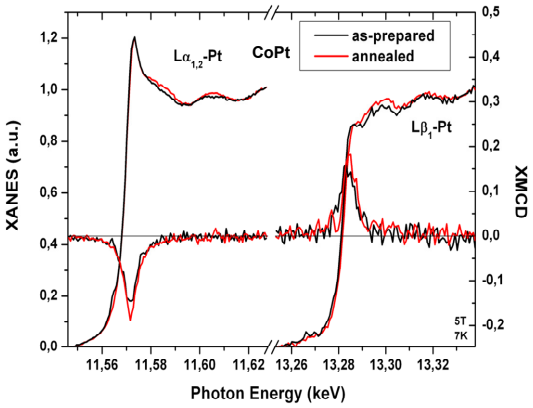
Experimental concerns

In the frame of this proposal, we performed XANES and XMCD measurements on as-prepared and annealed samples of binary FeAu and FePt 2 nm-particles embedded in a C matrix respectively at the Au and Pt $L_{2,3}$ edges. As a reference, we also studied the Pt edge EXAFS and XMCD on a mass-selected CoPt sample in the same condition to investigate the link between the spin-polarization of 3d-5d states. So the induced 5d magnetic moments were measured at a temperature close to 7 K and under magnetic fields up to 5 T. The magnetic moment per Au and Pt atom and the orbital to spin moment ratio were obtained by applying the sum rules. Our samples are very dilute (about 1 to 2 monolayers equivalent) within the matrix and the high brilliance of the ID12 beam line was essential: getting a reliable full XMCD spectrum at the

L_2 and L_3 5d edges took about 20 hours of acquisition time. All spectra were recorded in partial fluorescence yield using the 35-channel silicon drift detector.

Experimental results

First of all, from Pt-L edges extended x-ray absorption fine structure (EXAFS) experiments and simulations on our CoPt sample, we determined the first-shell Co-Pt and Pt-Pt coordination numbers and mean atomic distances and exactly retrieved the VASP *ab-initio* values expected for a 2nm $L1_0$–like phase cluster in agreement with our recent Co-K edge EXAFS experiments[1]. Then, X-ray magnetic circular dichroism (XMCD) investigations revealed a significant increase of both orbital and spin induced Pt magnetic moments for the annealed CoPt sample compared to the as-prepared sample (see figure and Table1).



Pt-L edge	m_S (μ_B/at.)	m_L (μ_B/at.)	m_L / m_S
CoPt as-prepared	0.38	0.038	0.1
CoPt annealed	0.53	0.09	0.17
FePt annealed	0.525	0.074	0.14

Figure 1*: XMCD spectra obtained on as-prepared and annealed CoPt sample at the Pt $L_{2,3}$ edges*
Table *1: Spin and orbital moments by applying the XMCD sum rules on CoPt and FePt samples at the Pt $L_{2,3}$ edges*

Nevertheless, while both the magnetic anisotropy energy (MAE) from SQUID measurements and the orbital contribution of annealed CoPt samples at Pt-edge are twice that of the as-prepared samples, the MAE value for such chemically ordered clusters is still one order of magnitude smaller that what is expected for the $L1_0$ CoPt bulk. Notice that the Pt L-edge XMCD measurements on the FePt sample, revealed that both orbital and spin contributions are of the same order of magnitude than for CoPt sample.

In the other hand, XMCD investigations on the annealed FeAu sample revealed limited induced Au magnetic moments compared to the FePt sample and to fcc FeAu disordered alloys[2]. This outcome could possibly be related to segregation effects. Finally, reversely to the Co-based sample, no relevant change is detected at 5d site between the as-prepared and annealed Fe-based samples.

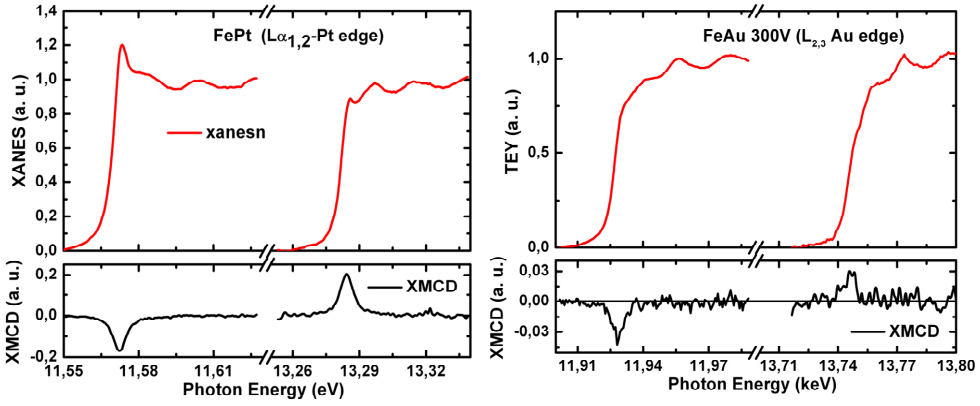


Figure *2: XMCD spectra obtained on annealed FePt (a) and FeAu (b) clusters at 5 d $L_{2,3}$ edges*

[1] **"Element-specific quantitative determination of the local order in CoPt alloy nanoparticles : Experiment and theory"** N. Blanc, L. E. Diaz-Sanchez, A. Y. Ramos, F. Tournus, H. C. N. Tolentino, M. De Santis, O. Proux, A. Tamion, J. Tuaillon-Combes, L. Bardotti, O. Boisron, G. M. Pastor and V. Dupuis *Submitted to* Phys. Rev. Letters (2012)

[2] **"Au and Fe magnetic moments in disordered Au-Fe alloys",** F. Wilhelm, P. Poulopoulos, V. Kapaklis, J.-P. Kappler, N. Jaouen, A. Rogalev, A.N. Yaresko, and C. Politis, *Phys. Rev. B.* 77, 224414 (2008).