ESRF	**Experiment title:** *Magnetic moments at Cu sites of high Tc superconductor thin films*	**Experiment number**: HE2525
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Report:

In conventional superconducting materials, superconductivity and magnetism are mutually exclusive phenomena. However, in recent years this has been shown to not always be the case e.g. URhGe [1]. In the High critical Temperature Superconductors (HTS) many theoretical models suggest that the pairing is mediated by antiferromagnetic fluctuations exchange interaction within the CuO_2 planes. Some recent investigations with neutron scattering [2], have also shown evidences of a presence of a "hidden" order parameter, predicted by Varma [3], associated to a patterning of the orbital moments due to circulating currents in the CuO_2 planes. Symmetry reasons make the detection of these feature very difficult, even using local probes like muon spin resonance and x-ray magnetic circular dichroism (XMCD). More recently, the existence of a magnetic internal field in optimally doped $YBa_2Cu_3O_{7+\delta}$ (YBCO) single crystals has been detected by Mossbauer spectroscopy on ^{57}Fe atoms substituted at Cu(1) site of the CuO chains [4]. The signature of a magnetic signal, attributed to the coupling between Cu and Fe atoms, has been interpreted as the experimental evidence of an antiferromagnetic interactions between the Cu ions.

In this experiment we have studied the X-ray Magnetic Circular Dichroism (XMCD) on copper of different high quality cuprate thin films. In previous experiment (HE2097) we already found the presence of a substantial paramagnetic moment on copper in superconducting $NdBa_2Cu_3O_{7-\delta}$ (NdBCO) very thin films (only 5 nm thick). In that experiment we found that the temperature dependence of this magnetic moment was strongly linked to the magnetic spin of the neighbor Nd rare earth. In order to elucidate the origin of the Cu-XMCD signal observed, we have studied the temperature dependence, the magnetic field dependence, and the effect of electric field doping, on the XMCD of YBCO, NdBCO and $La_{1.85}Sr_{0.15}CuO_4$ (LSCO) thin films. The characteristic of the samples studied are summarized in Table 1 together with the main outcomes. We found that in all the samples investigated an XMCD signal was detected, whose intensity was not obviously connected with the doping, with the film thickness or with the presence of magnetic rare earth in the compound. As shown below in details, the XMCD signal has a strong spin component, a negligible and strongly quenched orbital component, and is larger in the YBCO and NdBCO films, compared to LSCO. These results suggest that a strong contribution to the XMCD comes from the Cu(1) in the chains, and only a smaller part of the signal is related to the CuO_2 planes. However this analysis is subject of further theoretical investigation.

	Orbital Moment (μ_B/atom)	Spin Moment (μ_B/atom)	Number of Holes	Critical Temperature
YBCO 100 nm			0.15	88 K
Gracing Incidence	0.010	0.047		
Normal Incidence	0.008	0.038		
LSCO 50 nm			0.08	20 K
Gracing Incidence	0.002	0.01		
Normal Incidence	0.001	0.01		
NBCO 100 nm			0.10	60 K
Gracing Incidence	0.002	0.026		
Normal Incidence.	0.005	0.023		
NBCO 5 nm			0.06	20 K
Gracing Incidence	0.021	0.055		

Table 1: summary of the characteristics for the samples measured.

Experimental method

For YBCO, NdBCO and LSCO films we measured the XMCD signal using the high resolution Dragon monochrometor of ID08 and the 6T superconducting magnet at the $L_{2,3}$ edge of Cu. In this experiment the x-rays were tuned to the peak of the Cu $2p_{3/2}$ (L_3) and $2p_{1/2}$ (L_2) photo-absorption. The photon beam was at normal incidence to the sample (along the c-axis of the crystal lattice), and at grazing incidence (30 degree respect to the ab-plane). The spectra were measured for both light helicities (right and left). In normal and grazing incidence we used the total electron yield method, sensitive to the varying applied magnetic field and in particular at the interface, together with fluorescence yield method more to the bulk.
The measurements were performed as function of the temperature fixing the magnetic field at 4 T, whereas at low temperature the magnetic moment was analyzed changing the magnetic field.
We found a Cu magnetic dichroism in $YBa_2Cu_3O_{7-\delta}$ (YBCO) and $NdBa_2Cu_3O_{7-\delta}$ (NdBCO) films. Also on $La_{1.85}Sr_{0.15}CuO_4$ (LSCO) thin films a substantial XMCD was found, but strongly reduced compared to YBCO and NdBCO. The differences among YBCO, which contains two non-equivalent Copper sites (Cu(1) on the chains, and Cu(2) in the superconducting planes), and LSCO (containing only one CuO_2 plane and no chains), suggest an important magnetic contribution of Cu in the chains.
In figure 1 the absorption spectra and the XMCD [%] of YBCO film as function of the temperature is shown. The trend of the XMCD, confirm that at low temperature there is a magnetic moment on Cu. The XMCD decrease as a function of the temperature, but is still present above the superconducting transition. We used the sum rules to estimate the orbital and spin moment on Cu site. We found a strong contribution of the spin moment to the magnetic dichroism at the Cu site (0.06 μ_B/atom for NdBCO and 0.05 μ_B/atom for YBCO thin films at 9 K, see figure 2), while the orbital moment remains well below 0.01 μ_B/atom for all samples in the whole temperature range (Table 1). This in agreement with the know quenched orbital moment in cuprates. On the other hand the presence of a spin component, suggest that the spin associated to Cu, is not fully antiferromagnetically aligned, as it happens on the parent undoped compound.
This absolute value of the spin momentum observed here is similar to the values found in ref. [5] by Chackalian et al., that studied the interplay between magnetism and superconductivity in YBCO/LCMO multilayers. In ref. [5] it was concluded that the magnetic moment on Cu was induced at the interface by the presence of the neighbor ferromagnetic aligned Mn spins. Our results suggest, on the contrary, that the magnetic moment on Cu is not necessarily induced by the ferromagnetic layer.
The temperature dependence of spin moment as function of the magnetic field suggest a paramagnetic behavior in pure superconducting films. Indeed, as shown also in figure 2, it increases exponentially at low temperatures, and it does not have any obvious jump at Tc (in samples having different Tc’s).

As a consequence, the intensity of the XMCD signal is probably determined by the competition between thermal disorder and the magnetic field in the orientation of the canted Cu-spin. So at high temperatures, in our film due to the presence of holes, there is a spin canting effect, which give rise to the paramagnetic behavior. The paramagnetic behavior is confirmed by the magnetic field dependence of the XMCD signal studied at low temperature for all the samples studied. Again applying the sum rules, we find the both orbital and spin moment are linear as function of the intensity of the field. Non saturation, and no histeresys has been detected. It must be mentioned that the use of films with different thickness suggest that the results are not just a consequence of an interface effects.

The experimental results obtained are extremely interesting in view of the actual models of HTS superconductivity. Indeed the simultaneous presence of intrinsic magnetic moment on Cu and superconductivity may be an essential ingredient of the mechanism.

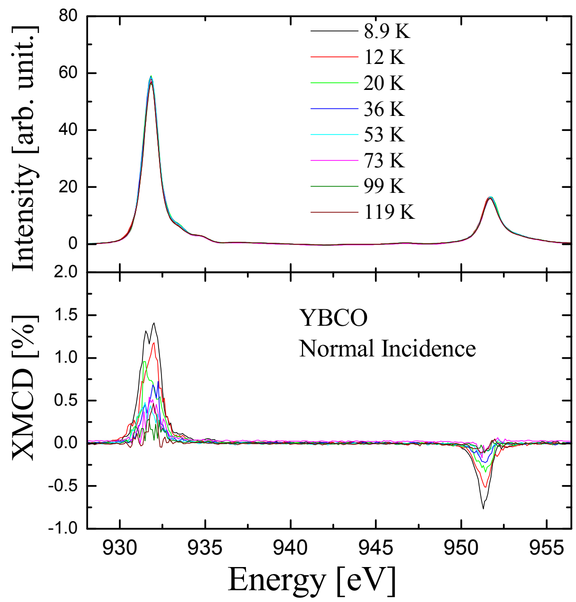

Figure 1: a) XAS spectra of the Cu $L_{3,2}$ of $YBa_2Cu_3O_{7-\delta}$ for different temperature, b) XMCD [%] obtained fromy the difference of the absorption spectra taken with different light helicity

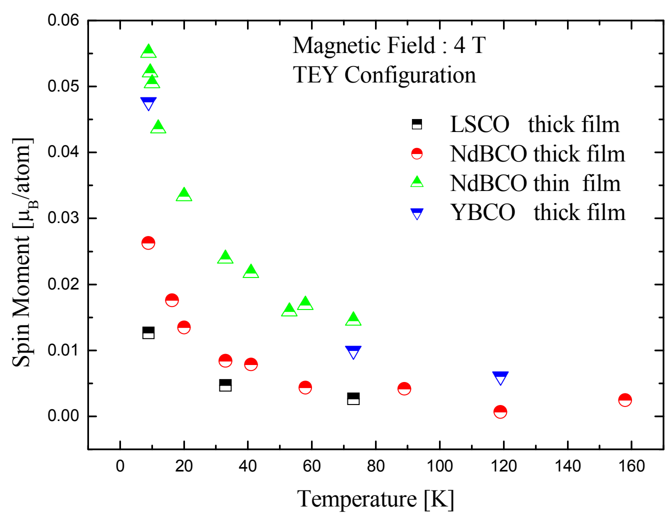

Figure 2: Spin moment as function of the temperature for LSCO, NdBCO thick and thin, and YBCO films, calculated with the sum rules from the XMCD spectra measured in total electron yield at 4 T

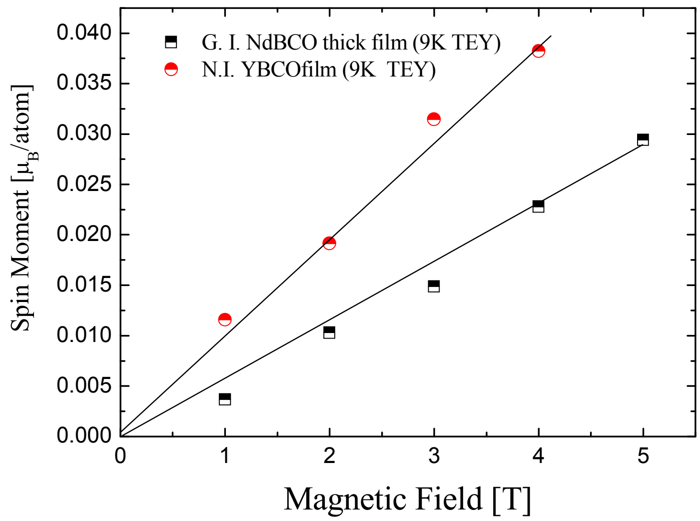

Figure 3: Spin moment as function of the magnetic field for YBCO and NdBCO 100 nm films, calculated with the sum rules from the XMCD spectra measured in total electron yield at 9 K.

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