

**Experiment title:**

Magnetic properties of Fe-atoms on CuN: Combining X-ray absorption with inelastic tunneling spectroscopy measurements.

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Report:

The aim of the project was to combine the information obtainable by the well established technique of XMCD with the new approach of spin excitation spectroscopy using scanning tunneling spectroscopy (SE-STs). This new technique allows to access local magnetic information of systems down to a single atom, however, it is so far limited to specific systems barely studied by other means. The aim of this beam time therefore was twofold. Firstly we intended to crosscheck the values obtained by the spin excitation spectroscopy and second to crosscorrelate the results of these two very different approaches to gain a complete picture of the system investigated. In two recent studies the properties of Fe and Co atoms on CuN/Cu(001) substrate have been investigated by SE-STs [1,2]. The measured STs spectra can be explained using a spin only description. Both systems show high magnetic anisotropy energies for the moment caused by the low symmetry of the chemical surrounding. The originally proposed project was on Fe atoms on CuN/Cu(100), however, we had considerable problems with a time evolution of the Fe-XAS signal presumably resulting from contaminations of the surface by the residual gas. In addition we have studied the magnetic properties of Co monomers on the same surface. The time effect, though present and hindering, was considerably smaller for this system. From the STs measurements it is predicted that the Co-moments have a strong magnetic hard axis along the Nitrogen rows (insert in Fig.1 a)). Due to the symmetry of the CuN surface there are two identical Co adsorption sites possible on the N-rows having the magnetic hard axis rotated by 90° with respect to each other (insert in Fig.1 a)). Since XMCD is a locally averaging technique, the two "species" of Co-atoms will

contribute with equal amounts to the spectra. In sum this results in an effective out-of-plane easy axis. From the x-ray measurements we find, first of all, that the spectra show sharp features and signs of a multiplet structure which points towards atomic like properties (Fig.1). This is in good agreement to the assumed weak interaction of the Co to the underlying Cu substrate, due to the insulating CuN monolayer, found by STS. We were not able to align the Co-moments parallel to the field at its maximum strength of 4.9 T as seen from the hysteresis shown in the insert of Fig.1 b). Therefore a direct determination of the magnetic moment and its spin and orbital contribution are not possible from the XMCD data. However, from the spectral shape we can conclude that the electronic state has a predominant d8-character [3]. Using a free atom description that would result in an spin of 1 while in the STS measurements a spin of 3/2 was found, however, this approach might be too simplified for this system. As shown in Fig.1b) the L2 edge has barely no XCMD signal, indicating a considerable orbital moment in the system. Even without knowing the saturation values we can estimate the ratio of orbital to spin moment (plus the dipolar term) to be about 2/3. This is not necessarily in disagreement to the STS measurements as the latter is expected to measure spin excitations that may, to first order, not be sensitive to orbital moments. We have also made a set of measurements with linear polarized light finding a clear anisotropy between the in- and out-of-plane Co-orbitals. We started a collaboration with G. Van der Laan trying to extract the precise electronic state including spin and orbital moment, by matching simulated XAS spectra to the complete set of measured data. In addition, as shown in Fig.2, we find that the projected spin moment at the maximum field reduces by about 50% when rotating the sample from 0° to 70° indicating a strong out-of plane anisotropy easy axis. We are able to fully reproduce this XMCD result using the Hamiltonian including magnetic moment and anisotropy found in the STS measurement, except for a small additional anisotropy term with the easy axis out of the plane. Such a small anisotropy is, however, in full agreement to the STS results. Beside the studies of magnetic properties of monomers we have also studied the effect of larger Co-coverage up to 15% of a ML (Fig.3). At the deposition temperature of 20 K, we can assume a statistical growth of clusters. For 1.5% coverage almost only monomers are present at the surface (>90%), while only 50% (34%) of the deposited Co will be monomers for 10% (15%) coverage. We find that surprisingly the XMCD signal reduces drastically for larger coverage. This is a clear sign for antiferromagnetic coupling of the individual magnetic moments in the Co-clusters, signatures of which have also been found in STS measurements. In conclusion, we have made the first comparison between the results obtained by spin excitation spectroscopy and by an established technique. The XMCD and SE-STs data are in excellent agreement on the anisotropies of Co-monomers on CuN/Cu(100). Deviation may exist on the electronic ground state and therefore the spin moment. Calculations are currently underway to resolve this issue.

[1] C.F. Hirjibehedin, et al., Science 317 (2007) 1199.

[2] A.F. Otte, et al., accepted in Nature Physics.

[3] P. Gambardella, et al., Phys. Rev. Let. 88 (2002) 047202.

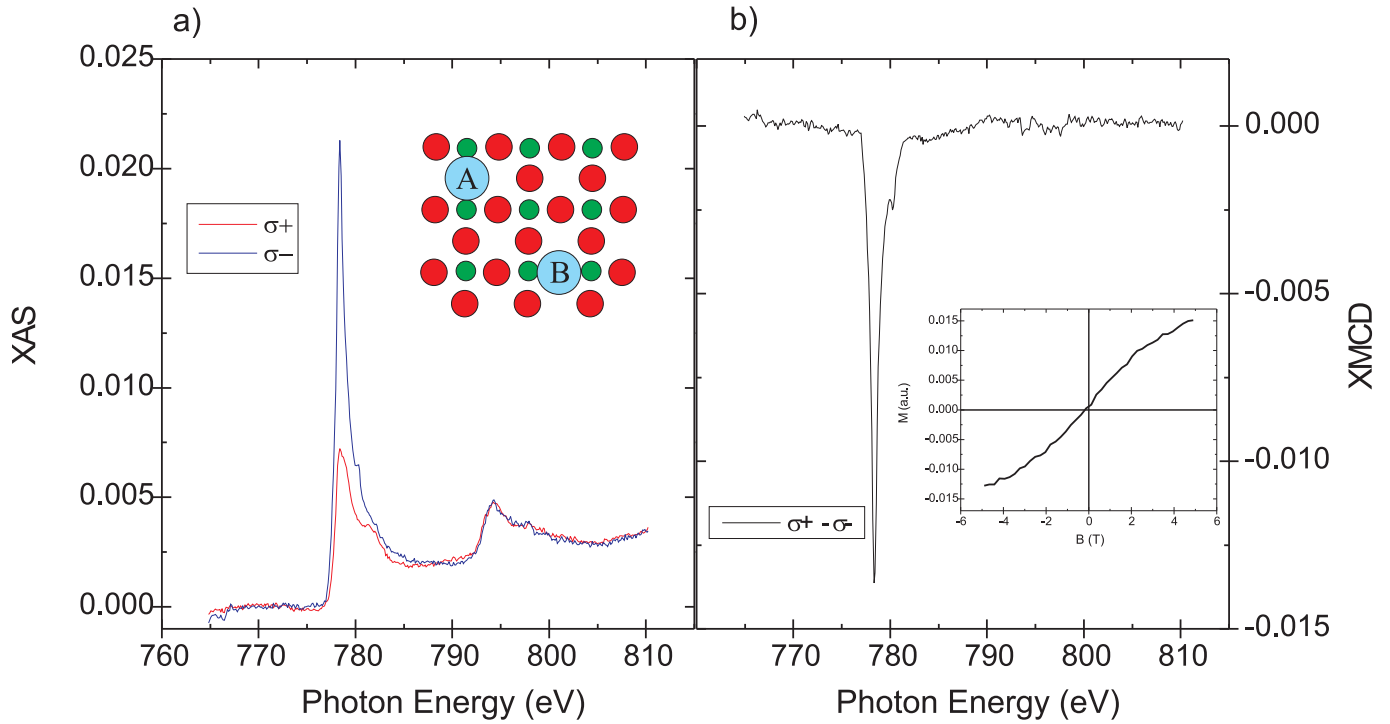


Figure 1: a) XAS spectra of 2% ML of Co on CuN/Cu(001) using circular polarized light. The substrate background has been subtracted from the spectra. The insert shows a scheme of the CuN surface (red: Cu, green: N) and the two possible adsorption sides of Co (blue) on the N-rows. b) XMCD data of the spectra shown in a) showing a clear d8-character. The hysteresis in the insert shows that it was not possible to saturate 2% ML of Co on CuN/Cu(001) with the maximum magnetic field.

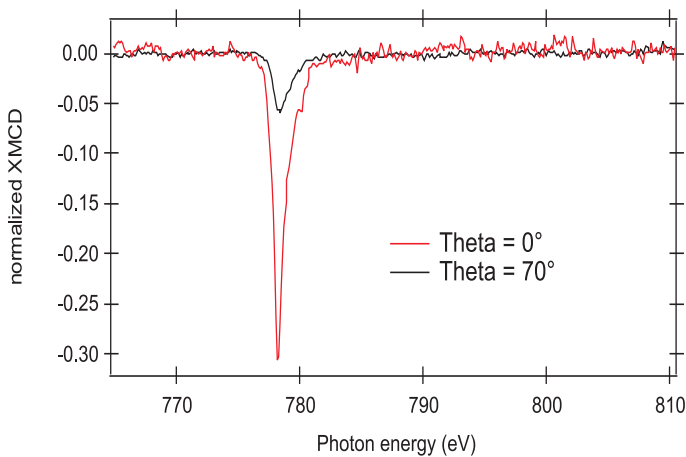


Figure 2: Angular dependence of the XMCD signal of 2% ML of Co on CuN/Cu(001). The spectra have been normalized to the integrated Co-XAS intensity and show a strong anisotropy with an out of plane easy axis.

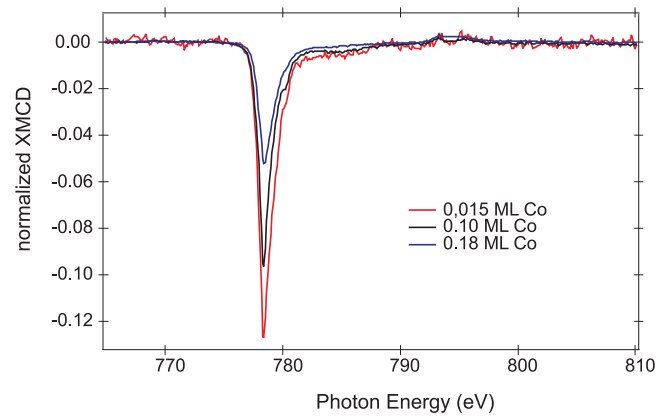


Figure 3: Coverage dependence of the XMCD signal of Co on CuN/Cu(001). The spectra have been normalized to the integrated Co-XAS intensity. Clearly the XMCD signal reduces substantially with the coverage indicating an antiferromagnetic coupling of the magnetic moments in dimers and larger clusters.

