J. Synchrotron Rad. (1999). **6**, 696–698

Magnetic *L*-edge EXAFS of 3*d* elements: multiple-scattering analysis and spin dynamics

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Magnetic EXAFS (MEXAFS) adds magnetic selectivity to the well established EXAFS technique opening the door to the exploration of local magnetic structure and disorder. Of particular interest is the behavior of 3d transition metal systems. By utilizing the enhanced $L_{3,2}$ MEXAFS signal, which is one order of magnitude larger than the K-edge MEXAFS, we performed a temperature dependent study of polycrystalline and epitaxially grown Fe and Co films. By analyzing single and multiple scattering contributions to the helicity dependent data, it is found that there are enhanced multiple scattering contributions in the magnetic case. In addition, we discuss the temperature dependence of the MEXAFS data which is linked to spin fluctuations.

Keywords: temperature dependent magnetic EXAFS

1. Introduction

The magnetic EXAFS (MEXAFS) is proving to be an important technique as one is able to probe the local magnetic structure. After Schütz *et al.* investigated the oscillatory fine structure of the difference of the X-ray absorption coefficients for right and left circular polarized light (Schütz *et al.*, 1989), the technique developed very fast. Today, the MEXAFS effect can be well described by theory (Brouder & Hikam, 1991; Ankudinov & Rehr, 1995; Brouder *et al.*, 1996; Ankudinov & Rehr, 1997; Ebert *et al.*, 1998). Therefore, one can now turn to more detailed investigations using MEXAFS. Although temperature dependent measurements provide vital information for magnetism, no such measurements had been performed for MEXAFS. The effect of enhanced multiple scattering contributions was discussed earlier (Ahlers *et al.*, 1998), however, a separation into multiple and single scattering contributions by comparison to theory was not performed. Here, we report EXAFS and MEXAFS measurements of 3d transition metals Fe and Co. As the magnetism of these metals is determined by the 3d-states, it is logical to measure at the $L_{3,2}$-edges as one uses p→d transitions. Moreover, the MEXAFS signal relative to the EXAFS signal at the L_3-edge is found to be about one order of magnitude larger compared to K-edge data (Ahlers *et al.*, 1998).

2. Experimental details and data analysis

The MEXAFS data have been recorded in two experimental setups. Epitaxially grown films of Fe (30ML) and Co (22ML) on a Cu(100) single crystal were measured at BESSY I (Lemke *et al.*, 1998; Srivastava *et al.*, 1998). Polycrystalline Fe (350ML) and Co (230ML) films were investigated at the NSLS (Wende *et al.*, 1998). The films were magnetized in plane and the angle of the photon wave vector $\boldsymbol{k}$ to the magnetization $\boldsymbol{M}$ was $\alpha(\boldsymbol{k},\boldsymbol{M})=20°$ for the epitaxially grown films and $\alpha(\boldsymbol{k},\boldsymbol{M})=45°$ for the polycrystalline films. The data presented here were normalized to $\boldsymbol{k}\|\boldsymbol{M}$ and full circular polarization ($P_c=1$). As shown earlier (Lemke *et al.*, 1998; Srivastava *et al.*, 1998; Wende *et al.*, 1998), the overlapping of the L_3 with the L_2-edge does not hinder the analysis. One does not find a quantum mechanical interference of the two contributions as the data are represented by the numerical sum of the two edges. That means that one can either investigate the directly extracted data and compare these to *ab initio* calculations where the L_3 and L_2 contributions are added (Lemke *et al.*, 1998; Srivastava *et al.*, 1998) or deconvolute the experimental data by an iterative Van Cittert analysis (Jones & Misell, 1970) and compare those to ab initio calculations of the L_3-edge only (Wende *et al.*, 1998). The L_1-edge jump was found to be 10% of the combined $L_{3,2}$-edge jump and has no pronounced features. Care was taken to eliminate the L_1-edge jump by the use of an appropriate spline function.

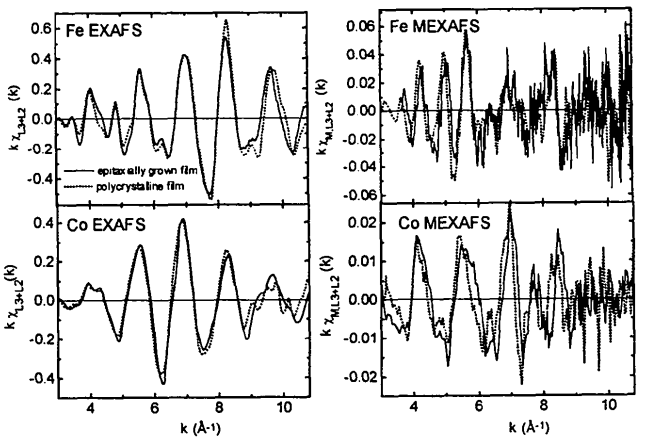

Figure 1
EXAFS and MEXAFS oscillations for Fe and Co as a function of the photoelectron wave number for the polycrystalline films (dotted line) and the epitaxially grown films (solid line).

3. Results and discussion

In Fig. 1, the oscillatory fine structure for the polycrystalline and epitaxially grown Fe and Co films are presented. Both for Fe and Co the EXAFS and the MEXAFS of the two experimental setups are very similar. That means that the local crystallographic structure and the local magnetic structure for the polycrystalline and the epitaxially grown film are the same. For the detailed analysis of the measurements the Fourier transformed data of the polycrystalline film are shown in the inset of Fig. 2. The data were recorded at four temperatures ranging from 70K to 400K. By comparing the Fourier transform of the normal EXAFS to MEXAFS we find the structures to be quite comparable. This is not very surprising as the local crystallographic structure and the local magnetic structure should be the same for a ferromagnet. By analyzing the intensities of the first peak of the MEXAFS in comparison to the normal EXAFS we find that the MEXAFS

intensity is about 5% of the EXAFS. This is about an order of magnitude larger compared to K-edge MEXAFS. One major difference between the normal EXAFS and the MEXAFS is the stronger temperature dependent damping for the MEXAFS. For a more detailed analysis of this effect the intensities of the first peak of the Fourier transform are analyzed. This is also shown in Fig. 2 where the reduced spontaneous magnetization is plotted as a function of the reduced temperature. The solid line represents a polynomial given in the literature (Stearns, 1986). In order to compare the intensity of the near edge magnetic circular X-ray dichroism (MCXD) signal with the Fourier transform intensity of the EXAFS and the MEXAFS, all the points of the lowest temperature are scaled to match the literature. As expected the temperature dependence of the MCXD signal follows the temperature dependence of the magnetization. The temperature dependence of the normal EXAFS signal exhibits a much stronger decrease. This can be explained by the thermal vibrations described by the Debye temperature of this film of θ_D =520K (Wende *et al.*, 1998). As the Curie temperature of Fe bulk is T_C =1050K, the decay of the EXAFS signal is much faster with temperature compared to the MCXD signal. An even stronger temperature dependence of the MEXAFS signal is a surprising result. In a simple picture one expects a similar trend for MEXAFS as for the normal EXAFS. In the normal EXAFS the thermal vibrations lead to a broadening of the pair distribution function. As a result the peak height in the Fourier transform decreases. This effect can also be expected for the MEXAFS signal as the spin dependent scattering potentials also change due to thermal vibrations. This would explain the same temperature dependence for the MEXAFS compared to the EXAFS.

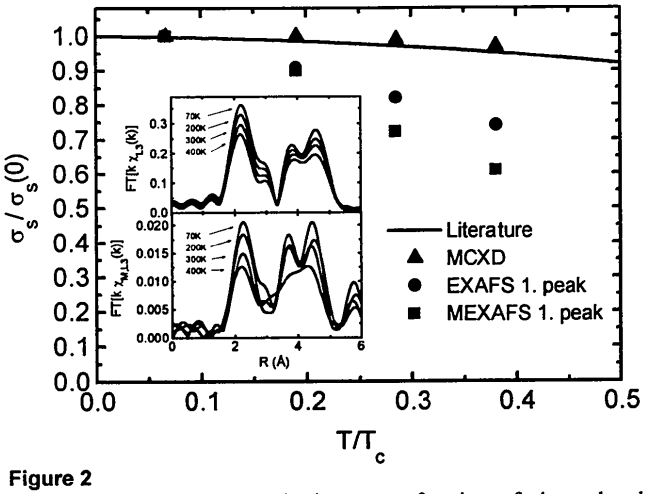

Figure 2
Reduced spontaneous magnetization as a function of the reduced temperature. The solid line is taken from the literature (Stearns, 1986). The inset shows the temperature dependence of the Fourier transformed data of the polycrystalline Fe film (upper part: EXAFS, lower part: MEXAFS).

But, we find an even stronger decrease for the MEXAFS signal (Wende *et al.*, 1998). This effect is not totally understood. We address the stronger decay to an additional disorder for the local magnetic structure. This disorder may be explained by additional spin fluctuations on a local scale. By comparing the relative intensities of the higher shells to the first peak in the Fourier transform, we find significant higher intensities for MEXAFS compared to EXAFS. In order to understand this effect we performed *ab initio* calculations using the FEFF7s code (Ankudinov & Rehr, 1997). The results are shown in Fig. 3. We find strong multiple scattering contributions for the MEXAFS which are not present for normal EXAFS (Lemke *et al.*, 1998). This can be seen by comparing the two calculations. The dotted line in Fig. 3 describes the contributions from single scattering only. The black line represents the calculation which includes multiple scattering and single scattering.

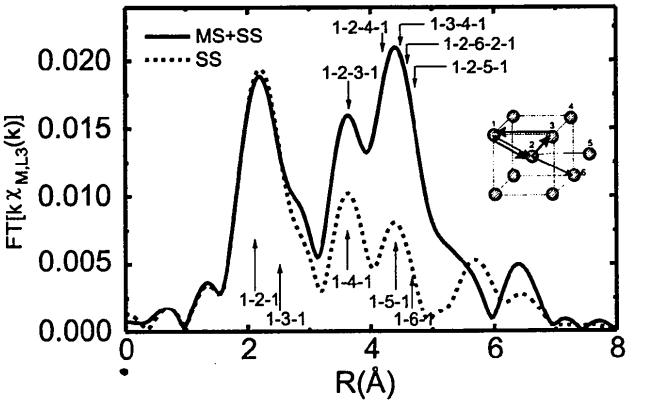

Figure 3
Ab initio calculation of the MEXAFS for the bcc structure using the FEFF7s code. The single scattering contributions (dotted line) have been separated from the combined multiple and single scattering contributions (solid line). The peaks are assigned to the different scattering paths which are labeled according to the inset.

For instance, the peak at about 3.6Å consists of the triangular scattering path described by 1-2-3-1 with about the same intensity as the single scattering path 1-4-1. A strong contribution of the peak at 4.4Å is due to the focusing path 1-2-6-2-1. This enhancement of multiple scattering paths was already discussed earlier (Ahlers *et al.*, 1998) but a separation of the paths which is a necessary proof of the effect was not presented.

4. Conclusion

We have presented MEXAFS measurements for polycrystalline and epitaxially grown Fe and Co film. The local crystallographic and magnetic structure of the polycrystalline and epitaxially grown film are quite comparable. The analysis of the temperature dependence shows that the MEXAFS signal is influenced by thermal vibrations. Moreover, as compared to EXAFS an even stronger temperature dependence for MEXAFS was found which is addressed to additional spin fluctuations. Comparison of the experimental data to *ab initio* calculations enabled us to separate multiple and single scattering contributions. Enhanced multiple scattering contributions were found for MEXAFS.

We want to thank N. Haack, G. Ceballos and A. Ney for their help during the experiments. This work was supported by the DFG SFB 290 and BMBF project No. 05 SC8 KEA.

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(*Received 10 August 1998; accepted 14 December 1998*)