Non-symmetric influences in the total electron yield X-ray magnetic circular dichroism signal in applied magnetic fields

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The total electron yield current is strongly influenced by external magnetic fields. As known before, this side effect can be slightly reduced by applied external bias voltages increasing the total sample drain current nearly up to saturation. Nevertheless those effects are not perfectly reduced in almost all X-ray Magnetic Circular Dichroism (XMCD) applications and are more prominent in very small XMCD signals, like O K edge spectra. We show that asymmetries in the total electron yield field response will result in XMCD offset signals, which are strongly photon energy dependent and follow the nonmagnetic absorption signal. A simple but effective method to prevent those offset signals, is the use of asymmetric magnetic fields. A quantitative analysis and a numerical reduction method for those offset signals are shown.

Keywords: total electron yield; x-ray magnetic circular dichroism; systematic errors

1. Introduction

X-ray Magnetic Circular Dichroism (XMCD) is the magnetization dependent absorption of circular polarized light in the vicinity of an absorption edge (Schuetz 1987). XMCD is now a well established method for site selective determination of spin and orbital moments (Altarelli 1993; Carra 1993; Chen 1994; Chen 1995; O'Brien 1994; Thole 1992). In the majority of soft XMCD experiments, below 2000 eV, the absorption coefficient is measured indirectly using total sample drain current or **T**otal **E**lectron **Y**ield (TEY) (Attenkofer 1997; Chen 1995; O'Brien 1994) or **F**luorescence **Y**ield (FY) (Goedkoop 1997). TEY has a significantly better intrinsic noise statistic, because the dominant decay channel of a produced core hole is the Auger process. Each Auger electron has a high scattering probability with other electrons in the system which results in a 'shower' of inelastically scattered low energy electrons leaving the sample. Therefore the absorption coefficient is nearly proportional, neglecting saturation effects (Chakarian 1997; Henneken 2000; Idzerda 1994), to the amount of excited core holes in a surface near region. In a XMCD measurement intrinsic **M** and extrinsic **H** magnetic fields are present. The trajectories of the low energy electrons leaving the sample are strongly affected by magnetic fields. This influence is usually not the same for different signs of magnetic fields (Goering 2000) and will result in an additional difference or offset signal in the XMCD measurement, which is not related to dichroism and can not be reduced by simply subtracting a straight line from the dichroism signal.

2. Experimental

We measured an epitaxial thin film of Fe on GaAs at the ESRF undulator beamline ID 12b, with an energy resolution of about $\Delta E = 0.2$ eV and a degree of circular polarization of nearly 85% using the 'flipper' magnet system with maximum magnetic fields up to ±5 kOe. Measurements were performed at a temperature of 10K (see Fig. 1 and 2).

A Fe(0.4nm)/Gd(0.4nm)x75 multilayer system was measured at the BESSY I SX-700 III beamline, with an energy resolution of about $\Delta E = 0.8$ eV and a degree of circular polarization of nearly 69% using a superconducting magnet system with maximum magnetic field of ±30 kOe. Measurements were performed at room temperature. The sample potential was held at -40V to minimize magnetic field influences and to increase the TEY signal. All data shown here were carefully corrected for dark currents, which are present with closed beamshutters.

All TEY signals are normalized to the photocurrent of a gold mesh avoiding synchrotron dependent intensity variations. No further smoothing was applied to all presented data. For all XMCD data points, the magnetic field was changed at each point by fixed photon helicity. The photon beam is coaxially aligned to the magnetic field.

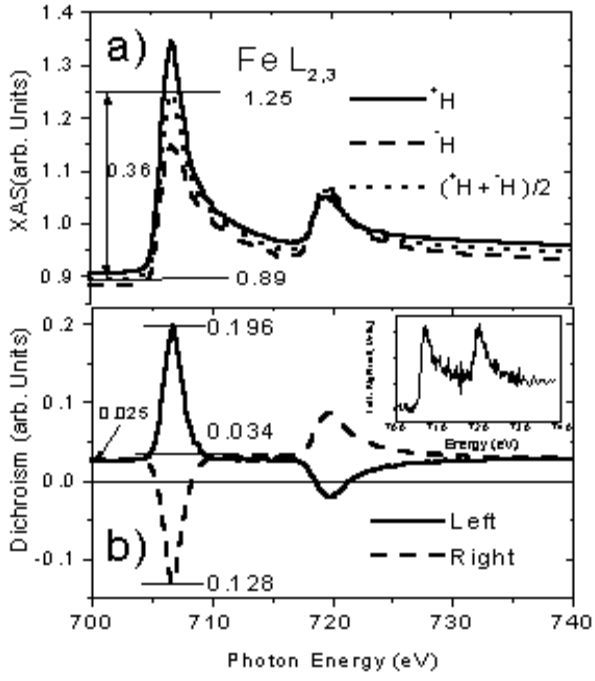

Figure 1: a) Fe $L_{2,3}$ TEY signal for parallel ($^{+}$H) and antiparallel ($^{-}$H) magnetic field of 4.3 kOe and both averaged TEY_{ave}. b) XMCD difference TEY signal ($^{+}$H- $^{-}$H) for left and right circular polarized light (inset shows XMCD sum over both helicities)

In Fig. 1a Fe $L_{2,3}$ Total Electron Yield spectra of a 8 ML Fe film grown on Ga As are shown and its corresponding XMCD (Fig. 1b) for two different light helicities. The magnetic field was flipped between ±5 kOe. The angle between the surface normal to the direction of the synchrotron light was 70°. The baseline of the dichroism signal is clearly shifted to positive values. To extract the nonmagnetic - not XMCD related - part in Fig. 1b, the sum of left and right circular polarized 'dichroism' signals is shown in the inset of Fig. 1b. This difference signal is quite comparable to the averaged TEY signal in Fig. 1a.

The field dependence of the TEY signal was measured at two different photon energies, one with no dichroism present (below the edge) and the other at the maximum of the dichroic effect of the L_3 absorption edge. The field dependent behavior below the edge is shown in Fig.2 . A strong and complex field dependancy is

observed. This behavior *f(B)* is not symmetric in external magnetic fields with opposite sign, which is indicated by the arrows at the left in Fig.2 . In our case the function values, in units of the normalized TEY signal, are *f*(- 4.3kOe*)* = 0.839 ≠ *f*(+ 4.3kOe*)* = 0.892.
From the ratio of 'before' to the 'on' edge field dependancy, a hysteresis loop could be extracted (Goering 2000), which is additionally shown in the inset of Fig.2 .

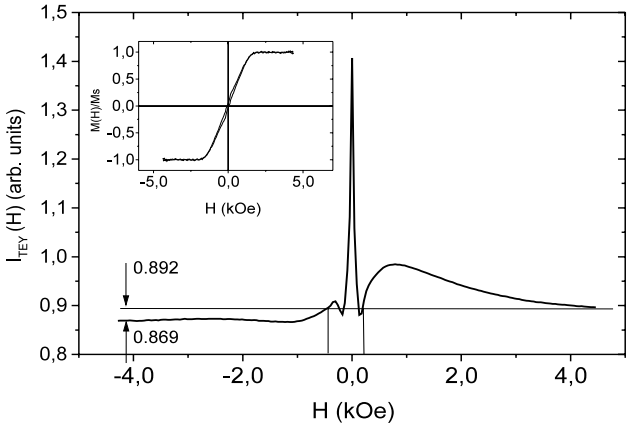

Figure 2 : Field dependent TEY signal below the edge of Fe (695eV) with no dichroism present; inset shows extracted Fe hysteresis loop

In addition we measured the magnetic field dependancy and the Fe XMCD hysteresis of an Fe/Gd multilayer system (Fig. 3). Using a symmetric external field of ±5 kOe. An offset has been observed in the XMCD signal at the Fe $L_{2,3}$ edges similar to Fig. 1 (not shown). We measured the XMCD signal with asymmetric field variation but with the same value of the *f(H)* function for positive and negative magnetic fields. From this functional behavior, shown in Fig. 3, we get the optimal XMCD-offset suppression fields at -4.05 kOe and +5.95 kOe with the smallest distance to the starting symmetric field variation of ±5 kOe. The resulting XMCD spectrum is shown in Fig. 4. The residual offset signal is smaller than 0.002. In a symmetric field variation the offset signal has been 0.2 below the edge. Looking at Fig. 3, this unsuppressed offset value can be easily extracted (dotted lines).
From our experience, we found that in a majority of experimental setups an asymmetry in the field dependance is present.

3. Discussion

The observed asymmetry in the total electron yield signal will directly influence XMCD related values, like spin- and orbital-momentum. For an easy explanation we start to artificially decompose the energy dependent absorption μ in terms of a nonmagnetic μ_0 absorption and a magnetization dependent absorption $\mu_c(M)$ and define:

$$\mu_{\pm} = \mu_0 + \mu_c(\pm M) \qquad (1)$$

Therefore the observed difference signal - shown in Fig. 1b - is $\mu_c(M) \cdot 2$. The measured TEY signal is nearly proportional, neglecting saturation effects, to the absorption coefficient and the proportionality is given by a field dependent function *f(H)*. The TEY signals for equal magnetized samples with different signs will be

$$I^+ = f(+H)\cdot\left[\mu_0 + \mu_c(+M)\right]$$
$$I^- = f(-H)\cdot\left[\mu_0 + \mu_c(-M)\right] \qquad (2)$$

Using $\mu_c(+M) = -\mu_c(-M)$ and calculating the difference signal gives

$$\Delta I = I^+ - I^-$$
$$= \mu_0 \cdot \left[f(+H) - f(-H)\right] \qquad (3)$$
$$+ \mu_c \cdot \left[f(+H) + f(-H)\right]$$

In the case of a symmetric field dependent TEY function *f(H),* this simply reduces to

$$\Delta I = \mu_0 \cdot 0 + 2 \cdot \mu_c \cdot f(|H|) \qquad (4)$$

This result is generally used implicitly in published XMCD TEY data. As shown in the experimental part, the function *f(H)* is often non-symmetric. Thus, the first term in equation 3 and 4 will not vanish and hence, an additional XAS signal is added to the XMCD signal.
For a quantitative analysis we assume, that no direct influence of

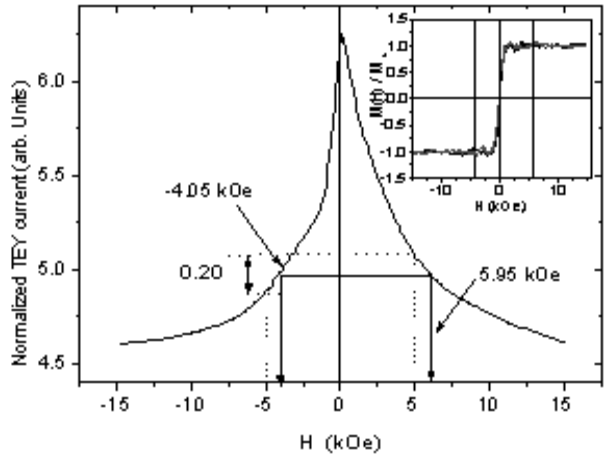

Figure 3: Field dependance of the TEY signal below the Fe edge; inset shows XMCD derived TEY hysteresis for the Fe L_3 edge

the helicity to the *f(H)* function is present. We start with the value below the edge in Fig. 1b of 0.025. This offset could also be extracted from the *f(H)* curve in Fig. 2 at identical external field values to 0.023. If the XMCD offset is only due to non-symmetric current behavior in *f(H)*, this energy dependent offset should be present in the whole XMCD spectrum. For quantitative proof, we look at the L_3 edge XMCD maximum, which is nearly located at the same photon energy compared to the white line maximum in Fig. 1a. The top of the left circular polarized spectrum has a maximal value of 0.196 and the bottom of the right circular polarized spectrum is -0.128. The center of gravity is at 0.034. If the offset signal is constant, this value should be 0.025, but this is not the case. To check the XAS related linear behavior of the offset at constant magnetic field, we calculate the offset at the maximum of the dichroic signal by multiplying the below the edge offset with the ratio of the XAS signal at the maximum XMCD to the below the edge value. Extracting values from Fig.1 a and b yields to 1.25 / 0.89 • 0.025 = 0.035, which is in perfect agreement to the center of gravity at the XMCD maximum and also consistent to the observed similarity of the XMCD sum for left and right circular polarized light in the inset of Fig. 1b.

For a numerical subtraction of the XMCD-offset, the XMCD spectrum *has* to be subtracted by the average XAS signal $TEY_{ave}(E)$ from Fig. 1a multiplied with the ratio 0.025/0.89.

Those offsets will influence the shape of XMCD spectra, which is directly related to integral values of the XMCD spectra and therefore the use of sum rules leads to wrong values. The influence to the orbital moment is usually strong, even for small residual offsets.
Using an asymmetric flipping field, same values for the field dependent TEY intensity function *f(H)* could be found in many cases. As an additional example, we show for a Fe/Gd multilayer system the typical *f(H)* dependency for negative and positive magnetic fields, measured with a superconducting magnet system at BESSY I and applied bias voltages up to -40V. The behavior below Fe L_3 edge is shown in Fig. 3. Looking at the XMCD hysteresis (inset in Fig. 3), a small reduction in the external field, does not change the saturated magnetization of our sample. Therefore we minimized the XMCD-offset, shifting the maximal and minimal field values by 0.95 kOe.

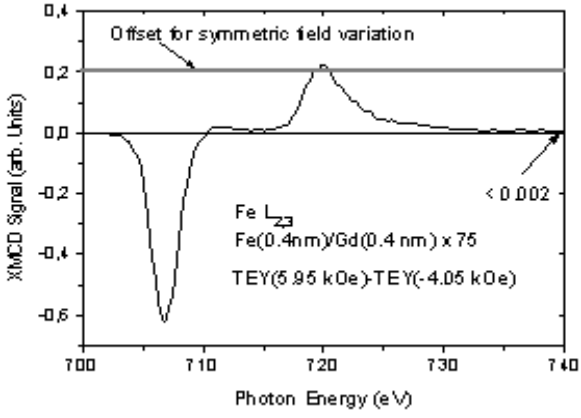

Figure 4: Fe $L_{2,3}$ XMCD, obtained with asymmetric field variation from -4.05 to +5.95 kOe. The below the edge offset for symmetric field variation (±5kOe) is additionally shown at an XMCD value of 0.2.

The measured XMCD signal is now nearly free of XAS-offsets. Residual offset values are about 0.002, which is 100 times smaller compared to symmetric field variations of 5 kOe (see Fig. 3).

For our first example at Fig. 2, an offset reduction using this asymmetric field variation is only possible at low magnetic fields, unfortunately with a corresponding reduction in sample magnetization (vertical lines at 0.3 kOe in Fig. 2). In such a case only numerical subtraction of offset signals is possible. The field averaged XAS signal has to be multiplied by a factor and subtracted from the XMCD signal to minimize the present offset.

An explanation for the complex behavior of the *f(H)* function is related to the magnetic field dependance of electron trajectories, which are spirals with field dependent radii. In applied fields a part of the electrons is able to move back to the sample surface. Hence, the corresponding TEY current can be decreased or changed. From this point of view we had simplified the field dependancy only to the external field. In a correct description the TEY signal depends on the external magnetic field and the sample magnetization *f(H,M)*. If the sample is perfectly flat and the experimental setup, like the sample holder, the sample itself, magnet, UHV chamber etc. are perfectly rotational symmetric around the photon beam spot and the photon beam itself has a cylindrical shape, no changes for opposite field directions should be present. This is hard to realize and in almost all experimental setups not possible. If the sample is quite large, the asymmetry could be reduced in many situations by moving the sample in some directions and minimizing the observed offset. For our first example (Fe on GaAs) this procedure could not reduce the offset signal in Fig. 1b. We do not believe that intrinsic mechanisms like spin dependent electron scattering or work functions are related to the observed offset features in the XMCD signal, because the majority of all the TEY electrons are secondary electrons, produced by inelastic electron-electron scattering, which are not spin polarized. If intrinsic mechanisms are related to this behavior, the sample position should not affect offsets in XMCD spectra, but it does.
In a TEY measurement additional offset signals with different character could be present, originating from electrons or ions inside the UHV system. Possible sources are ion getter pumps, vacuum gauges or stray light induced secondary electrons. Using the demonstrated numerical XMCD-offset reduction method, one has to check carefully the absence of those charge sources influencing the TEY signal.

4. Conclusions

We have investigated quantitatively the origin of typical offset signals present in XMCD TEY measurements. This offset is proportional to the nonmagnetic XAS TEY signal and it is described by slightly asymmetric field dependencies in the TEY current. We proposed a simple and effective method for numerical reduction of XMCD-offset signals present. Using asymmetric field variations, this offset could be reduced significantly during the measurement and no further subtraction is necessary.

5. Acknowledgement

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References

Altarelli, M. ; Physical Review B (1993) 47, 597 - 598
Attenkofer, K. and Schuetz, G. ; J. de Physique IV (1997) 7, 459 - 460
Carra, P., Thole, B. T., Altarelli, M. et al. ; Phys. Rev. Lett. (1993) 70, 694 - 697
Chakarian, V. and Idzerda, Y. U. ; J. Appl. Phys. (1997) 81, 4709 - 8979
Chen, C. T., Idzerda, Y. U., Lin, H. J. et al. ; J. Appl. Phys. (1994) 75, 6378 - 8979
Chen, C. T., Idzerda, Y. U., Lin, H. J. et al. ; Phys. Rev. Lett. (1995) 75, 152 - 155
Goedkoop, J. B., Brookes, N. B. et al. ; J. El. Spec. Rel. Ph. (1997) 86, 143 - 150
Goering, E., Fuss, A., Weber, W. et al. ; J. Appl. Phys. (2000), accepted
Henneken, H. et al. ; J. Appl. Phys. (2000) 87, 257 - 268
Idzerda, Y. U., Chen, C.T. et al ; Nuc. Inst. Meth. A (1994) 347, 134 - 141
O'Brien, W. L., Tonner, B. P. et al. ; J. Appl. Phys. (1994) 76, 6462 - 6464
Schuetz, G., Wagner, W. et al. ; Phys. Rev. Lett. (1987) 58, 737 - 740
Thole, B. T., Carra, P. et al. ; Phys. Rev. Lett. (1992) 68, 1943 - 1946