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Keywords
magnetic materials, gadolinium, X-ray absorption spectra, fine structure, iron, magnetic field effects, X-ray scattering

Disciplines
Condensed Matter Physics | Metallurgy

Comments

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On the Temperature Dependence of Multiple- and Single-Scattering Contributions in Magnetic EXAFS

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Abstract. We demonstrate that the temperature dependence of structural as well as magnetic fluctuations can be probed by the use of the Magnetic Extended X-ray Absorption Fine Structure (MEXAFS) spectroscopy. We compare those to the dynamic disorder as probed by the EXAFS. Here we present temperature-dependent MEXAFS investigations carried out at the L-edges of a thin Fe film and a Gd single crystal. By comparing the experimental results to *ab initio* calculations the single-scattering contributions are separated from multiple-scattering contributions. It is found that the multiple-scattering contributions are enhanced for the MEXAFS compared to the normal EXAFS.

INTRODUCTION

Today, the Extended X-ray Absorption Fine Structure (EXAFS) spectroscopy is a standard tool to in order to investigate local structure and dynamics. The importance of its spin-dependent counterpart – the so-called Magnetic EXAFS (MEXAFS) – has been recognized after the first wiggles in the extended energy range have been detected in 1989 [1]. Now, adequate literature exists on the theoretical description of the MEXAFS phenomena [2–4]. In the present work we demonstrate that a basic understanding of the spin-dependent electron scattering can be achieved by comparing *ab initio* calculations carried out using the FEFF7 code [2] with experimental data. We find an enhancement of the multiple-scattering contributions for the magnetic EXAFS compared to the normal EXAFS. The separation of the multiple- from the single-scattering paths can be easily carried out by 'turning off' the contributions of the multiple-scattering paths in the calculations. The use of this computational tool assisted us to develop a better insight into the spin-dependent electron scattering. Although temperature-dependent EXAFS measurements have been shown to provide essential information on the local dynamics, it is surprising that only few MEXAFS measurements have been carried out as a function of temperature [5–9]. Here we present temperature dependent MEXAFS measurements at the $L_{3,2}$-edges of a polycrystalline Fe film and at the L_3-edge of a Gd single crystal. After the separation of the multiple- from single-scattering paths, the temperature dependence of those contributions can be studied individually.

EXPERIMENTAL DETAILS

The Gd single crystal was investigated at the ID12A beamline of the ESRF. The crystal had the shape of a plate and was magnetized normal to the surface (hard axis). The measurements were carried out at normal x-ray incidence using fluorescence yield. The MEXAFS measurements of the polycrystalline Fe film have been carried out at the NRL facility located at the U4B beamline of the NSLS. The film was magnetized in-plane (easy axis). The data were taken at 45° x-ray incidence in transmission geometry. The comparison of the normal EXAFS of the polycrystalline film to data recorded for an epitaxially grown Fe film on a Cu(100) substrate revealed that the polycrystalline film is well ordered on a local scale [8]. We have shown earlier that the overlap of the L_3- with the L_2-edge does not hinder the MEXAFS analysis [5–9]. The data for the Gd and the Fe samples were taken in an applied magnetic field high enough for magnetic saturation as was determined by means of element-specific hysteresis loops.

RESULTS AND DISCUSSION

We start with a qualitative discussion of the temperature dependence of the MEXAFS and EXAFS oscillations at the Gd L_3-edge. The experimental data are presented in Fig. 1 for two temperatures (10K and 250K). The upper panel shows the EXAFS wiggles which exhibit a clear temperature-dependent damping. Using a correlated Debye-model we determine a Debye-temperature of θ_D=155K. This justifies the strong reduction of the EXAFS wiggles at 250K $> \theta_D$ since one expects an exponential attenuation via the EXAFS Debye-Waller factor $e^{-2\sigma^2 k^2}$. Here, σ^2 is the mean square relative displacement. The exponential damping is seen clearer when the intensity decrease at low k-values is compared to the damping at higher k-values. At k=4.5Å^{-1} the EXAFS signal at 250K is reduced to 69% of the 10K value whereas at k=10.0Å^{-1} the signal decreases to 16%. A similar damping of exponential form is found for the MEXAFS. This indicates that the thermal vibrations influence the magnetic EXAFS signal as well. Furthermore, there is at least an additional damping for the MEXAFS as the temperature increases due to the reduction of the magnetization.

We have observed such effect for Gd, Fe and Co [5–9]. This indicates that such effects are general in nature and are not linked to the specific electronic structure of the material. In order to discuss the different aspects of the temperature-dependent

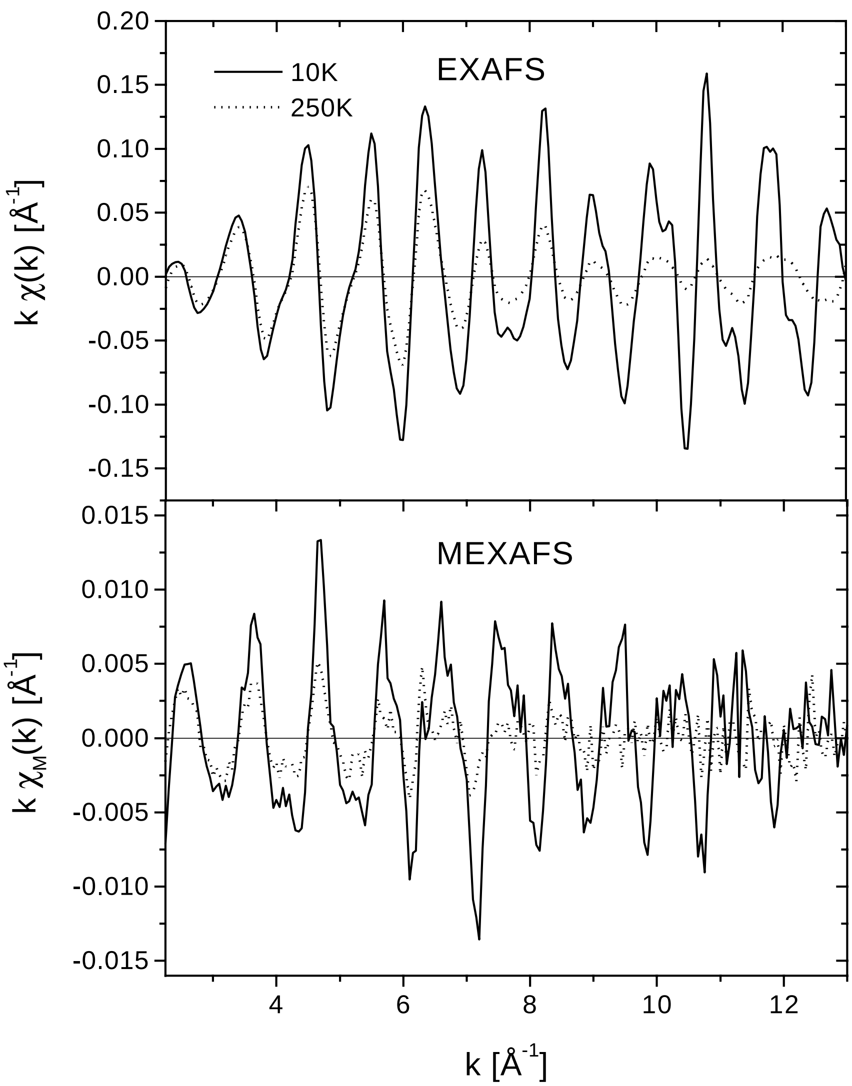

FIGURE 1. Temperature dependent Gd EXAFS $k\chi(k)$ and MEXAFS $k\chi_M(k)$ taken at the L_3 edge.

damping of the EXAFS and the MEXAFS in more detail, we now turn to a quantitative discussion for the EXAFS and MEXAFS of a polycrystalline Fe film. Before investigating the temperature dependence, it is useful analyze the Fourier transformed EXAFS and MEXAFS data at the lowest temperature. For a better understanding of the contributing scattering paths theoretical calculations were carried out using the *ab initio* FEFF7 code [2]. The results are presented in Fig. 2 where the Fourier transform of (a) the EXAFS and (b) the MEXAFS oscillations are shown. In both cases the dotted lines represent the results using single-scattering paths only. The calculations including single- and multiple-scattering contributions are given as solid lines. These calculations combining multiple- and single-scattering are in good agreement with the experimental data as can be seen in comparison to the inset of Fig. 3 and in Refs. [7,8]. The main peak of the EXAFS as well as the MEXAFS is determined by the nearest (1-2-1) and the next nearest (1-3-1) neighbor single-scattering paths. Comparing the calculation for the second and the third peak in the Fourier transform of the EXAFS and the MEXAFS a clear enhancement of the multiple-scattering contributions can be determined for the magnetic case. The triangular scattering path 1-2-3-1 contributes about 20% to the intensity of the second peak for the normal EXAFS whereas a contribution of about 40% is found for the MEXAFS. The enhanced multiple-scattering is also found for the third peak. Here numerous multiple-scattering paths are identified. The strongest contribution is due to the focusing path 1-2-6-2-1. For the third peak of the MEXAFS Fourier transform the multiple-scattering paths dominate the single-scattering ones, as about 65% of the peak intensity are due to multiple-scattering. A contribution of 50% only is found for the normal EXAFS. The enhancement of the multiple-scattering paths for the MEXAFS can be described in a phenomenological picture as discussed in Ref. [10]. The effect of the exchange interaction is introduced into the scattering process by means of a spin-dependent scattering amplitude F_M. This is scaled by the spin-polarization $\langle\sigma_z\rangle$ and is added to the Coulomb scattering amplitude F_0. Therefore, the backscattering amplitude becomes $F = F_0 \pm \langle\sigma_z\rangle \cdot F_M$ for right and left circularly polarized light, respectively. The backscattering amplitude for a multiple-scattering path of n scattering events can then be approximated to:

$$(F_0 + \langle\sigma_z\rangle \cdot F_M)^n \approx F_0^n \cdot \left(1 + n \cdot \langle\sigma_z\rangle \cdot \frac{F_M}{F_0}\right) . \quad (1)$$

Thus, the multiple-scattering contributions can be enhanced for the MEXAFS by the factor n compared to the normal EXAFS.

Since the individual contributions to the Fourier transform peaks have been identified by means of the *ab initio* calculations, we now turn to the analysis of the experimental results. The temperature-dependent Fourier transforms of the experimental EXAFS and MEXAFS oscillations for the polycrystalline Fe film are presented in the inset of Fig. 3. A clear temperature-dependent damping can be seen for both cases. It was shown earlier that the lattice vibrations which determine the damping of the EXAFS are well described by a Debye-Temperature of θ_D=520K [7] analyzing the first EXAFS peak. An even stronger temperature-dependent damping is found for the first MEXAFS peak. We try now to correlate this damping to the magnetic part of the disorder. We show in Fig. 3 the reduced spontaneous magnetization as a function of the reduced temperature together with the intensity of the near edge Magnetic Circular X-ray Dichroism (MCXD) signal and (a) the first, (b) second and (c) third EXAFS and MEXAFS Fourier transform intensities. For

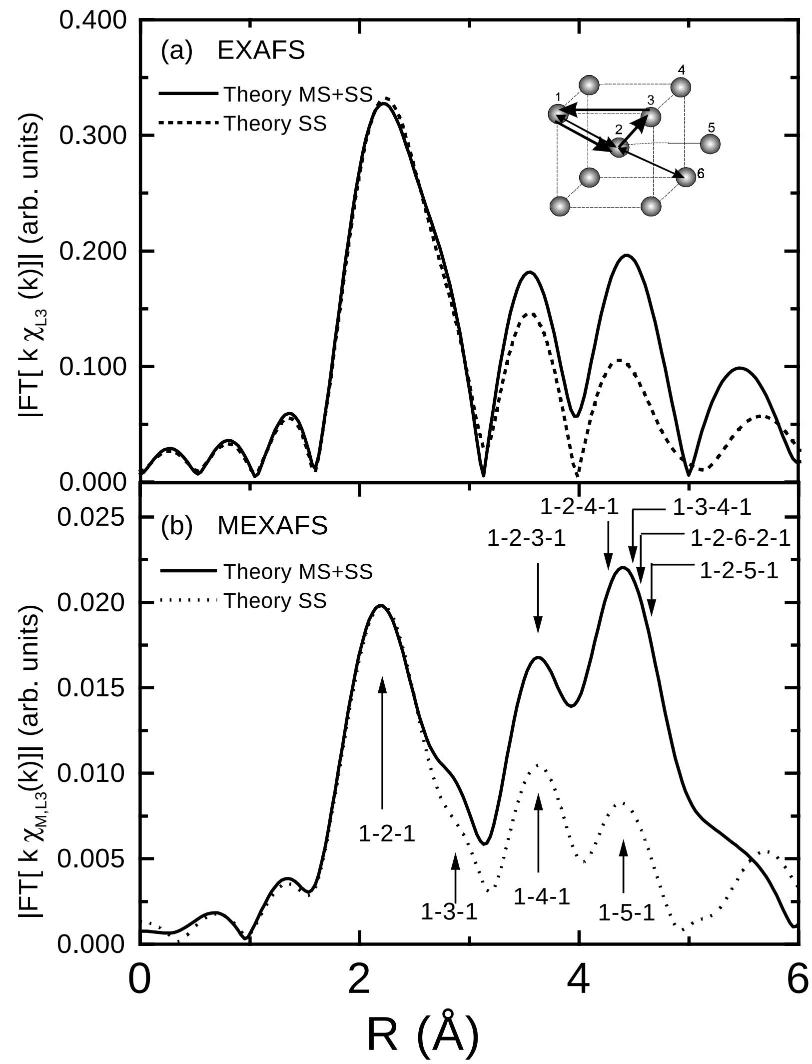

FIGURE 2. *Ab initio* calculation of Fourier transforms $|FT[k\chi_{L3}(k)]|$ and $|FT[k\chi_{M,L3}(k)]|$ of (a) the EXAFS (b) and MEXAFS oscillations for Fe bcc structure at 70K using the FEFF7 code. The single-scattering contributions (dotted lines) are separated from the combined multiple- and single-scattering contributions (solid lines). The peaks are assigned to the different scattering paths which are labeled according to the inset.

a relative comparison, the MCXD as well as the EXAFS and MEXAFS intensities are scaled to match the literature at the lowest temperature. The MCXD signal follows the temperature dependence of the magnetization as shown in Fig. 3(a).

This is not the case for the first EXAFS and MEXAFS peak of the Fourier transform. First we discuss why the EXAFS peak exhibits a much stronger temperature dependence. The temperature dependence of the normal EXAFS is determined by the Debye-temperature of θ_D=520K. Therefore a much stronger damping is

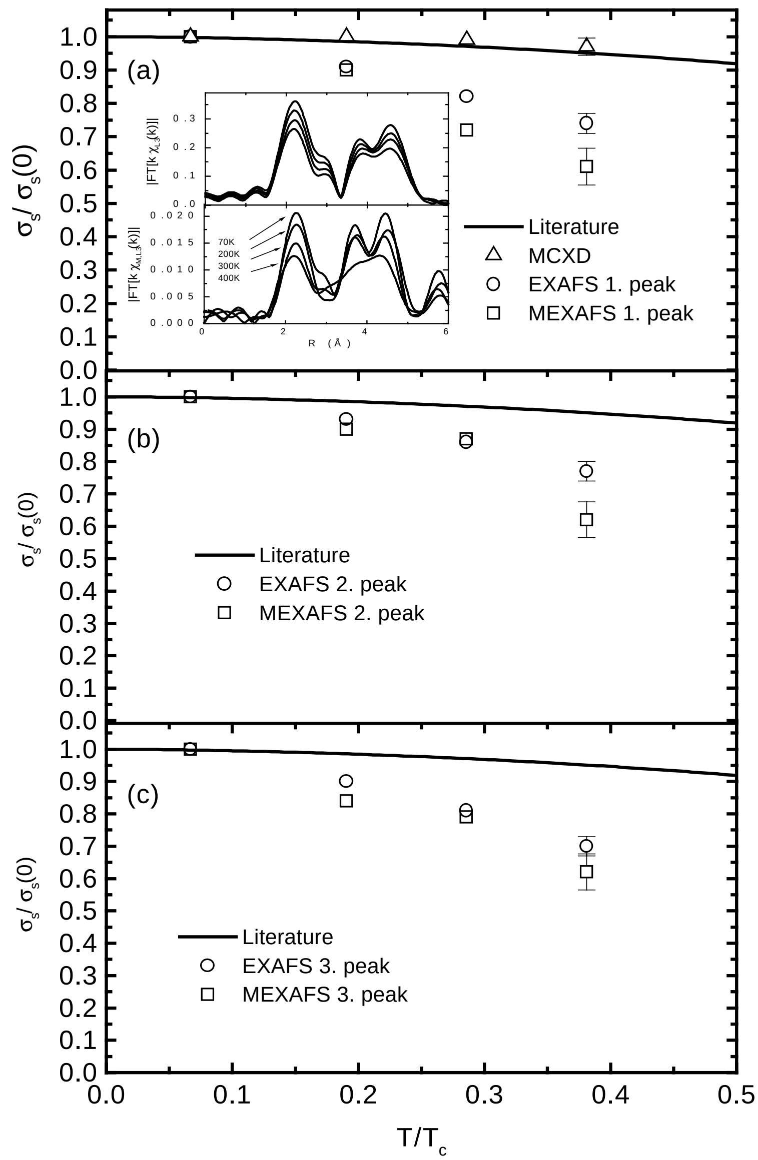

FIGURE 3. Temperature dependence of the EXAFS and MEXAFS Fourier transform intensities for the polycrystalline Fe film. The reduced spontaneous magnetization is given as a function of the reduced temperature (taken from the literature [11]). The Fourier transform intensities are scaled to match the literature values at the lowest temperature. The inset shows the temperature dependence of the Fourier transforms for the experimental EXAFS (top) and MEXAFS (bottom) oscillations.

found at T=400K (which corresponds to a reduced temperature of T/T_C=0.38) in comparison to the reduction of the MCXD signal which is determined by the Curie-temperature of T_C=1050K. This is due to the fact that up to a reduced temperature of $T/T_C \approx 0.3$ the attenuation of the MCXD signal is described by a $1-\beta T^{\frac{3}{2}}$ law. This shows a much smaller temperature-dependent decrease than the exponential damping which describes the normal EXAFS. A surprising result is therefore the even stronger temperature dependence of the first MEXAFS Fourier peak in comparison to the EXAFS signal (Fig. 3(a)). At a temperature of 400K (T/T_C=0.38) the MCXD signal is reduced to only 97% of the T=0K value whereas the EXAFS signal is reduced to 74%. This shows that a simple multiplication of those values leading to 72% does not describe the observed damping of the MEXAFS of 61% with respect to the T=0K value (Fig. 3). This indicates that there is a larger magnetic disorder on a local scale (probed with the MEXAFS) compared to the long-range spin fluctuations (probed with MCXD) leading to the decrease of the magnetization. This difference in the "probing length" of the MCXD and the MEXAFS technique is due to the fact, that the mean free path of the scattered photoelectron is a function of the kinetic energy of the electron and differs for both cases. In the near-edge energy range (MCXD) the mean free path tends to diverge and therefore a long-range order is probed. In contrast, the MEXAFS probes the magnetism on a nearest neighbor length scale due to its more localized scattering origin and the fact that the mean free path exhibits a minimum of about 7Å at k=3.0Å^{-1} and increases to about 30Å at k=13.0Å^{-1}. Up to now we discussed the first peak of the Fourier transform which includes single-scattering contributions only. The second and third Fourier transform peaks contain strong multiple-scattering contributions for the MEXAFS case as can be seen in Fig. 2. This can be the origin of the observed anomaly in the damping of the MEXAFS signals around T/T_C=0.18, where a slower intensity decrease is observed (see Fig. 3 (b) and (c)). It is known that multiple-scattering contributions exhibit a stronger temperature-dependent damping. Therefore the damping of the multiple scattering paths will be seen mostly at lower temperatures, whereas the damping of the single-scattering paths will become effective at higher temperatures. Indeed for the second as well as the third peaks the same trend of a stronger temperature-dependent damping of the MEXAFS in comparison to the EXAFS is found in Fig. 3 (b) and (c) around T/T_C=0.4. These observations allow us to set boundaries to the temperature range in which each of those two mechanisms is mostly responsible for the damping.

CONCLUSION

We have presented two temperature-dependent magnetic EXAFS studies. For the Gd single crystal as well as for the polycrystalline Fe a clear temperature-dependent damping for the normal EXAFS and the magnetic EXAFS were determined. The quantitative analysis for the polycrystalline Fe film shows that thermal vibrations also influence the MEXAFS signal. The comparison of the damping of the normal EXAFS with the MEXAFS demonstrates that there must be a higher local magnetic disorder compared to the long-range order probed with MCXD. *Ab initio* calculations carried out with and without multiple-scattering contributions clearly indicate an enhancement of multiple-scattering paths for the magnetic EXAFS. This separation enabled us to discuss the temperature dependence of the individual multiple- and single-scattering paths.

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