

Local-spin-selective X-ray absorption: a new technique to study correlated materials

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Abstract

K edge X-ray absorption spectral shapes can be detected by measuring the $K\beta$ X-ray emission decay channel. Due to the coherence of the excitation and decay processes, the convolution of the spectral shape with the 1s life time broadening disappears. The increased resolution makes possible the study of electron correlation effects in the pre-edge peaks of K edges. Due to the 3p3d exchange interaction, the $K\beta$ decay channel has separate peaks for spin-up and spin-down. This spin-selection is exploited in so-called local-spin-selective X-ray absorption. From its comparison with X-ray magnetic circular dichroism (X-MCD) it is possible to determine the energy dependence of the Fano factor. This is shown for MnP.

1. Introduction

Local-spin selective X-ray absorption has been exploited for the first time by Hämäläinen and co-workers [1]. In this technique the K edge X-ray absorption spectrum is measured by detecting the $K\beta$ X-ray emission ($3p \rightarrow 1s$) decay channel at two different energy positions. One can separate the $K\beta$ X-ray emission spectrum into an internally referenced spin-up part and a spin-down part. This is shown in Fig. 1. The $3p \rightarrow 1s$ decay is calculated for divalent manganese. The spin-up (dashed) and spin-down (dotted) peaks are clearly separated. Details of these multiplet calculations are given by Peng and co-workers [2]. The influence of charge transfer effects on the $3p \rightarrow 1s$ decay is discussed in Ref. [3]. The energy window of the fluorescence detector can be tuned to the desired parts of the spectrum and in this manner one is able to

obtain spin selectivity in the K edge X-ray absorption spectrum. The use of a core spectrum for the spin selectivity implies that the spin is referenced to the local situation in the solid. For a ferromagnet this is equivalent to an external referenced technique, such as X-MCD.

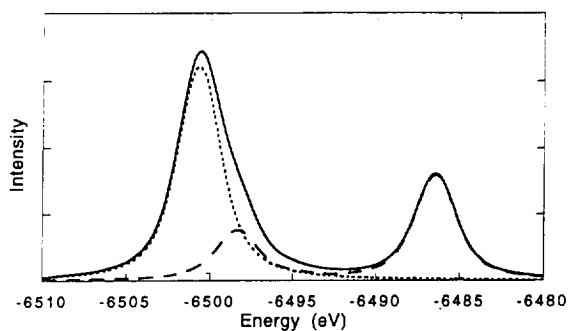


Fig. 1. Crystal field multiplet calculation for Mn^{2+} . The total spectrum of the $3p \rightarrow 1s$ decay is given with the solid line. It is separated into spin-down (dotted, main peak) and spin-up (dashed).

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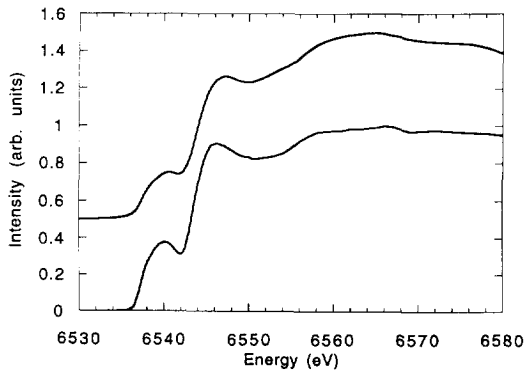


Fig. 2. K edge X-ray absorption spectrum of MnP. Top: the spectrum measured with transmission. The high-resolution spectrum measured at the peak position of the $K\beta$ decay is given at the bottom.

2. High resolution K edge X-ray absorption

Fig. 2 shows a comparison of the K edge X-ray absorption spectra of manganese phosphate (MnP) measured with transmission and with the detection of its $K\beta$ decay channel. Details of the experiments are described elsewhere [4]. The $K\beta$ detected spectrum shows increased experimental resolution [5]. For K edges it can be shown that the intensity of the $K\beta$ channel is given as the X-ray absorption cross section, multiplied respectively by a Lorentzian broadening of the 3p hole, the experimental broadening and the $K\beta$ cross section, which is a constant at a particular detection energy. As a rule K edges are described using single particle models, that is the excited electronic states are identified with the empty p-density of states. The high-resolution $K\beta$ detected X-ray absorption spectra promise to reveal new details of the electronic structure of solids. For example, for the $L_{2,3}$ edge of rare earths the high-resolution structures at the edge have clearly been identified as quadrupolar transitions [5]. A particular field which will be helped with high-resolution data is the interpretation of the pre-edge structures in iron compounds, for which many different models have been suggested [6]. New high-resolution X-ray absorption results have the promise to largely advance our understanding on this matter.

3. The comparison with X-MCD

For ferromagnetic materials there exist two probes to measure the spin-polarisation of the empty states:

X-MCD and local-spin selective X-ray absorption. In X-MCD one uses circularly polarised X-rays which are sensitive only to the orbital moments. One is able to detect the spin-polarisation due to the spin-orbit coupling of the final states. This gives rise to a proportionality factor P , the so-called Fano factor [7,8]. The Fano factor is determined by the relative size of the radial matrix elements for the final state with $j = 3/2$ and $j = 1/2$. It can be shown that for differences less than 5% the Fano factor is proportional to the difference in these matrix elements: $P = 2/3(R_{3/2} - R_{1/2})$. The ratio between X-MCD and local-spin selective X-ray absorption is a direct probe of the Fano factor [4].

Fig. 3 shows the energy dependence of the Fano factor for MnP. The zero energy is the X-ray absorption edge. For energies over 10 eV the statistics of both measurements did not allow a direct determination of P . The values averaged over 10 eV are given. From this comparison it is clear that the Fano factor, thus the difference in radial matrix elements for $j = 3/2$ and $j = 1/2$, is strongly energy dependent. At the edge, the Fano factor is approximately -4%. It decays quickly to values lower than 1% at energies above 10 eV. At higher energies the Fano factor seems to oscillate, however as yet the statistical and systematic errors in the measurements are too large to confirm this with certainty [4]. The assumption that the Fano factor is constant in energy is clearly refuted for values close to the edge. The origin of this strong energy dependence is related to the presence of the spin-polarised 3d-electrons at these energies. They induce an increased spin-polarisation for the p-electrons at

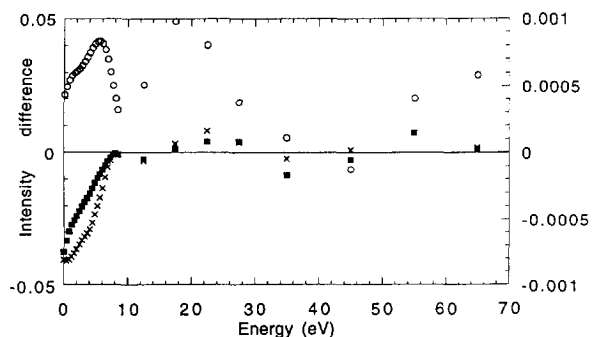


Fig. 3. Intensity differences normalized to the edge jump are given for X-MCD (crosses, scale on right) and local-spin selective X-ray absorption (open circles, scale on left). Their ratio, identified with the Fano factor, is given with solid squares (scale on left).

these energies which, apparently, strongly affects the radial matrix elements for $j=3/2$ and $j=1/2$.

4. Concluding remarks

For K edge X-ray absorption spectra, a considerably better resolution can be obtained with the detection of the $K\beta$ decay channel. These high-resolution X-ray absorption spectra promise to reveal many new details of the electronic structure of solids.

By comparing X-MCD and local-spin selective X-ray absorption we determined a Fano factor of -4% at the edge. Its value decreases quickly and at energies 10 eV above the edge it has decreased to values lower than 1%.

Acknowledgements

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