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ESRF

Experiment title: Quadrupolar transitions in cubic crystals	Experiment number: HE-323
Beamline: ID12A	**Date of Experiment:** from: Aug 22 1998 to: Aug 24 1998
Shifts: 6	**Local contact(s):** A. Rogalev, J. Goulon
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Report:

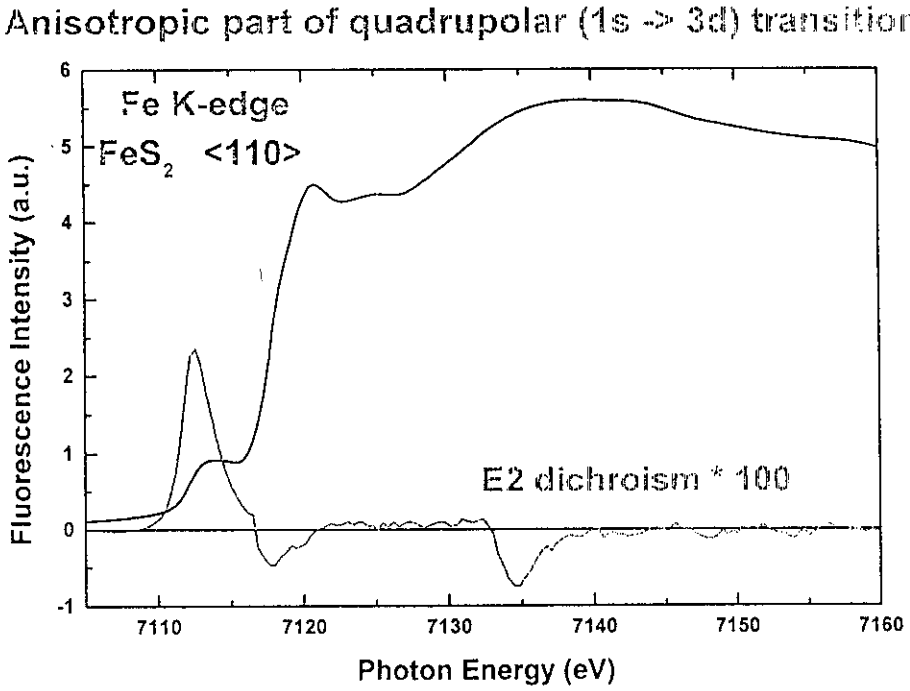
The purpose of this experiment was the investigation of quadrupole transitions in cubic FeS_2. The single crystal has been cut along a (110) face and glued on the sample holder. J. Goulon has implemented a new method to measure angular dependent spectra in single-crystals, which proved to be very convenient for pyrite. The sample was turned around an axis parallel to the x-ray beam and, at each energy, the fluorescence intensity was measured every 3 deg. In principle, the angular dependence should be a periodic function with period $\pi/2$. The average is the average absorption and the peak to peak amplitude represents the angular dependence. In practice, the angular dependence is much more complicated because of the presence of diffraction peaks in the single-crystals. The advantage of the measurement method is that diffraction peaks can be spotted easily as non-periodical peaks, and discarded. Then, a fit of the rest of the angular dependence as $a\cos 2\theta + b\sin 2\theta + c$ gives the average absorption c, the intensity and the phase of the angular dependence. This protocol is used at each energy point to deliver an angular dependent spectrum.

With this method, a very sensitive and accurate measurement of the angular dependence is possible. The angular dependent quadrupole transitions were found to be mainly in the pre-edge structure at the K-edge of iron. Its maximum intensity is half a percent, which is in agreement with the calculation carried out with a multiple-scattering program. The experiment was repeated on the two opposite faces of the sample, and identical results were obtained.

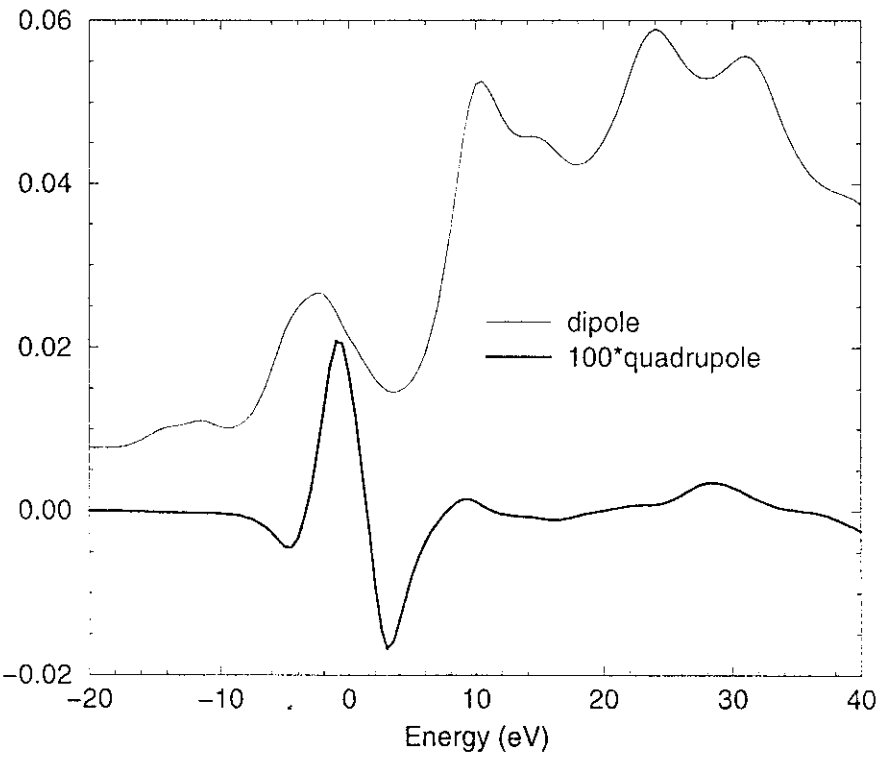
The experiment has brought two results. Firstly, it has shown that quadrupole transitions can be unambiguously observed at the K-edge of iron. Their intensities and positions can be measured accurately. The angular dependence of quadrupole transitions in pyrite have been calculated within the multiple-scattering approach and the multiplet theory. Only the multiple-scattering approach agrees with experiment, the multiplet theory overestimates strongly the angular dependence. This is important because it enables us to estimate the role of quadrupole transitions in XMCD spectra.

Secondly, it has shown that very unreliable results are obtained if diffraction peaks are not avoided. The usual method to measure angular dependent spectra in single crystals is to measure a spectrum, turn the sample and measure another spectrum. We have shown that this is a quite dangerous procedure, because the presence of diffraction peaks is not always clear. Sometimes, it is sharp "glitch", but it can also be rather smooth, and could be interpreted as an angular dependence. This could explain some results found in the literature.

Non-muffin-tin multiple-scattering calculations are now in progress to reproduce quantitatively the observed spectra.



Experiment
(preliminary results)



Multiple-scattering
calculation