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Report:

We have performed high resolution resonant inelastic X-ray scattering (RIXS) measurements on the molecular based magnet $CsNi[Cr(CN)_6]$. The synthesis of well characterized molecular based magnets with Curie temperatures close to room temperature remains a challenge where the competition is high [1]. Adjusting molecules and metals makes it possible to fabricate magnets with new properties, i.e. insulating, low density, low anisotropy, transparency, biocompatibility and magneto-optical sensitivity.

We measured the $K\beta$ emission of Ni around the Ni K absorption edge. Using conventional techniques to measure X-ray absorption, like transmission or total electron yield, the absorption spectra of deep lying core-levels are significantly broadened by the fast decay of the core hole. Hämäläinen and coworkers [2] have shown that one can get partly rid of this life-time broadening if the coherent decay is measured using a decay channel involving a shallower core level. In our case, the lifetime broadening of the Ni $1s$ level is replaced by the one of the $3p$ final state core hole of the $K_\beta(3p \to 1s)$ emission. In Figure 1(a) we report the K-edge absorption spectrum taken at the maximum of the K_β emission. The quadrupolar $(1s \to 3d)$ transitions are clearly discerned, as well as the different dipolar $(1s + np)$ contributions. K_β emission spectra taken at several photon energies around the different absorption features put into evidence the localized or itinerant nature of the different final states, like it was also done for the quadrupolar and dipolar contributions to the $Gd L_3$ edge [3].

Another application of RIXS is the use for local spin-selective X-ray Absorption Spectroscopy (LSXAS) [4]. Because of the exchange interaction between the $3p$ and $3d$ electrons, the K_β emission of Ni is split into a main peak and a satellite. The main peak, at lower energy, consists mainly of transitions leaving a spin-down hole in the $3p$ shell (anti-parallel to the majority spin in the 3 d band), while the satellite consists purely of spin-up transitions [5]. The difference between RIXS spectra taken with the analyzer energy fixed at the main peak and at the satellite can therefore in principle give information about the

local spin polarization of the measured element [4]. Since it is a local probe, long-range magnetic order is not necessary and this technique can also be used for anti-ferromagnetic and paramagnetic materials. For ferromagnetic compounds, a comparison between LSXAS and X-ray Magnetic Circular Dichroism (XMCD) can give the energy dependence of the Fano factor, as has been shown by part of the present team [6]. However, the spectrum taken at the satellite includes also a contribution due to resonant Raman effects and moreover the energy resolution is not the same, effects for which it is not easy to correct. In Figure 1(b) we present two spectra taken at different energies at the satellite, where the difference in spin-polarization is not optimal, but the influence of resonant Raman contributions is similar and thus easier to correct for. The agreement with the XMCD for the same compound [7] is clear, and the effect seems much larger due to the exclusion of the Fano factor which for Mn was evaluated to be a few percent [6]. However, there are some effects which might influence the obtained results. The first one is that we observed changes in the absorption spectrum, even taken at the same analyzer energy, due to the degradation of the sample by the high intensity of the incoming X-rays. A second effect is that the shape of the K_β emission changes as a function of the absorption energy due to broadening and contraction of the main line. Further experiments are necessary to completely understand and apply the LSXAS measurements on these compounds.
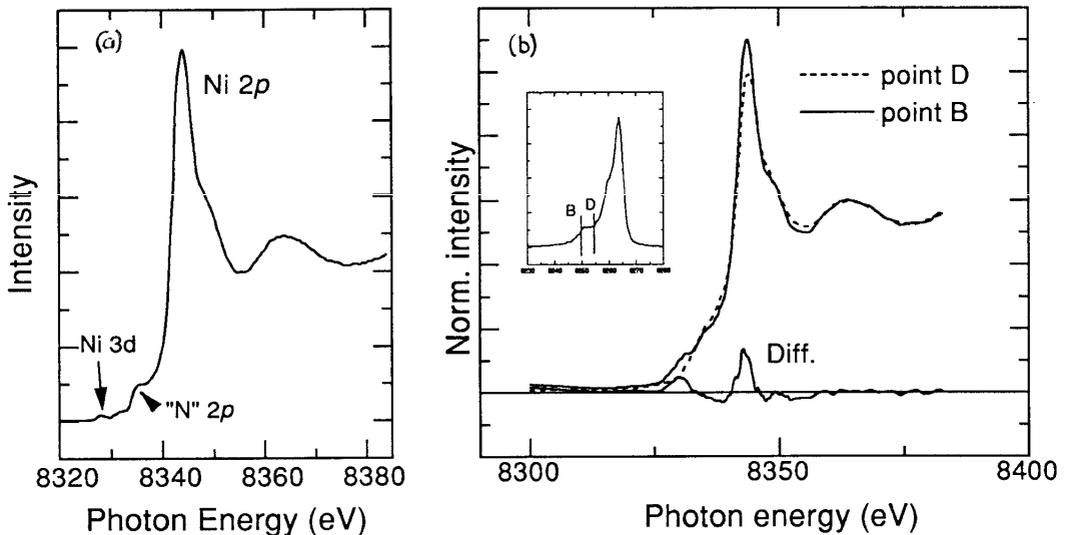

FIG. 1. (a) Resonant inelastic X-ray spectrum at the Ni K-absorption edge using K_β emission. The transitions of the 1s electron to the different final states are denoted in the spectrum.
(b) RIXS spectra taken at different analyzer energies, and difference curve.

In conclusion, using RIXS we have been able to put in evidence the contribution of $1s \rightarrow 3d$ quadrupolar transitions to the K-edge absorption spectrum of Ni in the molecular based magnet $CsNi[Cr(CN)_6]$. Absorption spectra taken at different energy positions at the K_β emission line show differences which are similar to XMCD curves obtained on the same compound. A more quantitative analysis, including a detailed comparison with theory, is being performed at the moment.

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