ESRF	**Experiment title:** Orbital magnetism of transition-metal impurities in noble metal hosts studied by XMCD	**Experiment number:** HE 1309
Beamline: ID 08	**Date of experiment:** from: 21.11.2002 to: 26.11.2002	**Date of report:** 25.02.2004
Shifts: 15	**Local contact(s):** P. Bencok	*Received at ESRF:*

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Report:

Direct Observation of Orbital Magnetism in Dilute $3d$ Impurities

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Abstract

We present X-ray magnetic circular dichroism (XMCD) determinations of the orbital/spin magnetic moment ratios of dilute $3d$-series impurities in Au (and Cu) host matrices. This is the first direct measurement of spin and orbital moments in dilute alloys, which represent an intermediate case between nearly-free-atom behavior and bulk transition metals or compounds. The relative importance of orbital magnetism, expressed by the moment ratio μ_l / μ_s, differs from that found in bulk materials, especially in the case of AuCo, where a large enhancement of the orbital moment is observed. The observed trends are in agreement with *ab initio* calculations of the spin and orbital moments of dilute $3d$ impurities in noble metals.

Submitted to Physical Review Letters, Nov. 2003

Figure 1 shows measured X-ray absorption spectra (XAS) for four dilute samples of $3d$ impurities in Au host, and the XMCD spectra derived from them. The large background from Au presented no serious problem and could be corrected for by subtraction of a background registered from pure Au in the same wavelength range. The corresponding atomic multiplet structures are shown as lines in the XAS spectra.

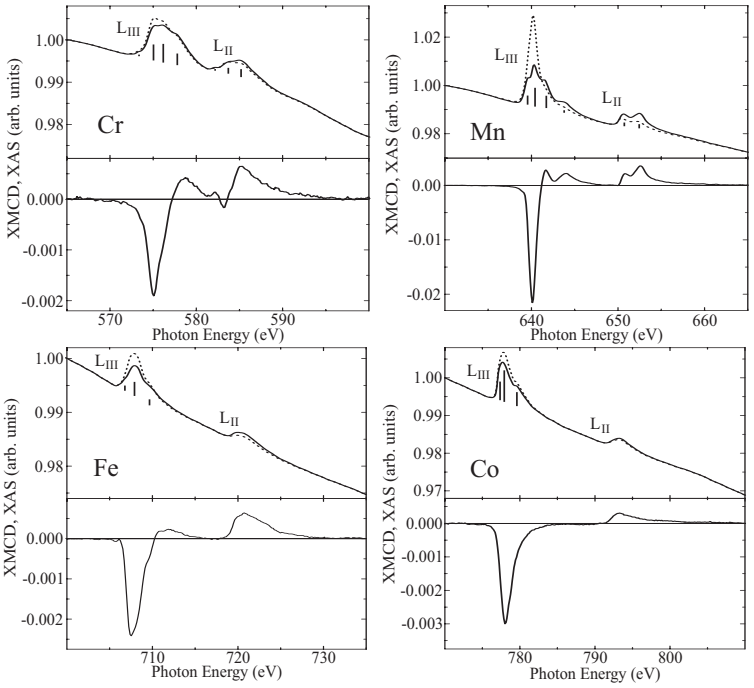

Fig. 1. XAS spectra (upper plots) and difference (XMCD) spectra (lower plots) of the 3*d* elements Cr–Co in the $L_{II,III}$ spectral region. Note the strongly reduced relative intensity at the L_{II} edge of Co, indicative of its enhanced orbital moment.

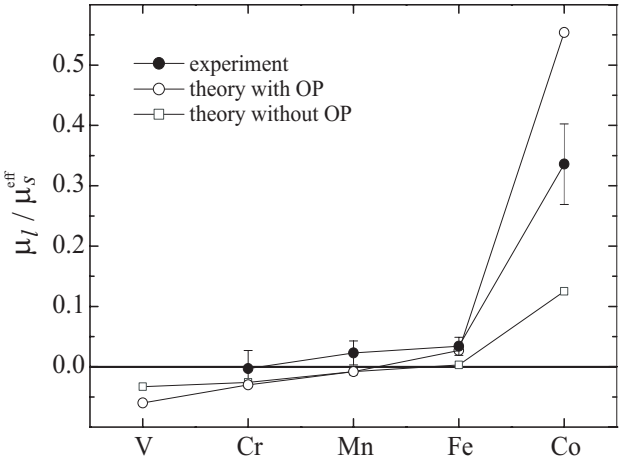

Fig. 2. Systematics of the orbital/effective spin moment ratio for the 3*d* series as impurities in Au host. The experimental results from XMCD (filled points) are compared with theory, with (open circles) and without (open squares) the orbital polarization (OP) correction. The true ratio μ_l / μ_s lies within the error limits shown.

Figure 2 shows the orbital/spin moment ratios derived directly from the spectra in Fig. 1 without a sum-rule analysis. They are compared with theoretical *ab initio* calculations and reproduce the calculated trends. Especially notable is the large orbital moment of Co, in agreement with the theoretically predicted trend.

In Fig. 3, $L_{II,III}$ XMCD spectra are shown for Mn dissolved in a Cu and a Au matrix, illustrating the influence of the host matrix on the local electronic structure. The spectra were normalized to unity at the L_{III} edge in order to emphasize the effects of the host. The overall line shape of the XMCD (and XAS) signal persists; however, the structural details are sharpened for Mn in Au. This can be understood by considering the stronger hybridization of the impurity with Cu, favored by its smaller lattice constant than in the Au host. Moreover, one sees a relative enhancement of the L_{II} XMCD signal for Mn in Cu. The resulting ratio μ_l / $\mu_s^{\rm eff} \approx 0$ shows that the stronger hybridization of Mn with Cu leads to a smaller value of the orbital moment in the Cu host.

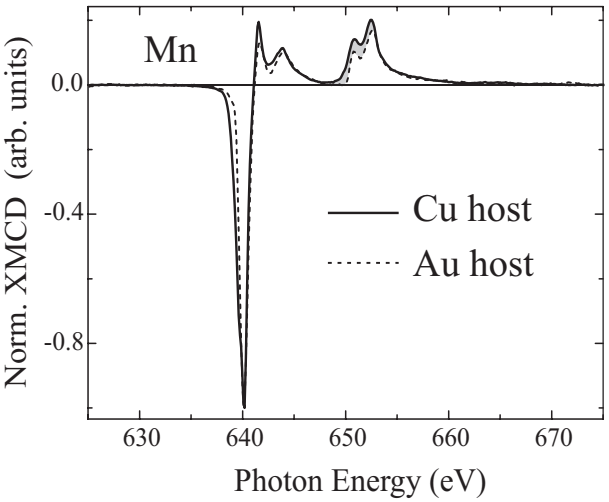

Fig. 3. $L_{II,III}$ XMCD spectra of Mn as a dilute impurity in Cu (solid curve) and Au (dashed curve) hosts. The ratio of peak areas in the L_{II} and L_{III} regions yields the orbital/spin moment ratio.

In conclusion, notable orbital contributions to magnetic moments of 3*d*-series dilute impurities in noble metal hosts have been directly observed for the first time using XMCD. The trend seen in going along the 3*d* series is in agreement with *ab initio* theoretical predictions, in particular the large orbital contribution for Co impurities, which is nearly four times the (already considerable) orbital moment ratio of Co metal. This is in strong contrast to Fe, where the orbital moment is reduced in Au host, and emphasizes the importance of details of the band structure in the appearance or suppression of orbital magnetism.