ESRF	**Experiment title:** **Magnetic moments of 3*d* impurities in alkali metal and noble gas hosts**	**Experiment number**: HE 991
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Report:

Since the work of Friedel, dilute transition metal systems in non-magnetic hosts constitute one of the most interesting fields in solid state physics. Aim of our proposal HE 991 (May 2001) was to disclose the electronic structure and local magnetization of diluted transition metal impurity systems by employing x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD). In particular, we intended to investigate the origin of the giant magnetic moments observed for 3d impurities in alkali metal hosts. We report two significant results obtained in our experiment. First, the present measurements demonstrate that XAS and XMCD have a great potential to study dilute systems with impurity concentration as low as $3x10^{12}$ atoms cm^{-2} providing simultaneous and direct information on both electronic and magnetic configuration of the impurity states. Second**,** by combining XAS and XMCD, we directly determined for the first time the valence state of Fe, Co, and Ni impurities in alkali metal hosts. The multiplet structure of the XAS spectra indicates that Fe, Co, and Ni have localized atomic ground states with predominantly d^7, d^8, and d^9 character, respectively. XMCD shows that the localized impurity states possess large, atomic-like, magnetic orbital moments that are progressively quenched as the isolated atoms form clusters. These data settle unambiguously the origin of giant magnetic moments observed for 3d metal impurities in alkali hosts that has been recently the object of an intense debate in the solid state community [1-7].

The experiment took advantage of the high radiation flux at the ID8 beamline and of the high magnetic field (7 T) and variable temperature range (10-300 K) available at the experimental station. Fe, Co, and Ni were evaporated in minute quantities (0.002 – 1 ML) on a K film prepared on a clean Cu(111) substrate. XAS at the $L_{2,3}$ edges was performed in total electron yield mode using circularly polarized light in magnetic fields up to 7 T with the sample at T=10 K. XMCD was recorded by switching both the light polarization and the sample magnetization. Figure 1 shows the XAS spectra recorded for Fe, Co, and Ni impurities deposited on a

K film for parallel (solid lines) and antiparallel (dotted lines) alignment of the light polarization with respect to the applied field. The XMCD spectra are also shown at the bottom of each panel. For each transition metal there are several notable features. The XAS spectra present narrow multiplet structures which are not observed in the corresponding bulk metal spectra (see Fig. 2a). This is a clear indication of 3d localization on the transition metal impurities. In addition, the magnitude of the XMCD is significantly larger compared to the bulk and even compared to that reported for low-dimensional structures, where the size of the magnetic moments are increased due to the reduced coordination [8]. Further, in the case of Fe and Co, the XMCD at the L_2 edge has the opposite sign with respect to bulk spectra, whereas it is zero for Ni (see Fig. 2b for Co). To determine the electronic configurations of the localized states, the multiplet structure of the XAS and XMCD spectra is compared to atomic multiplet calculations reported by van der Laan and Thole [9] for the $3d^n \rightarrow 2p^5 3d^{n+1}$ absorption thresholds. For zero values of the crystal field and for atomic spin-orbit splitting there is a very close match between the experimental XAS and XMCD spectra reported in Fig. 1 for Fe and Co and those calculated for d^7 and d^8 atomic configurations. For Ni ions no calculated d^9 spectra exist in the literature. The latter, however, is a straight forward case since the single peak at the L_3 edge indicates that the only states which are excited in the absorption process are those arising from the j=3/2 initial state. Dipole selection rules, in this case, imply that the empty d states are of purely j=5/2 character indicating a d^9 atomic ground
state with S=1/2, L=2, and J=5/2. The atomic ground states derived from our data are in very nice agreement with recent local spin density calculations [6,7]. Fe, Co, and Ni possess corresponding atomic-like magnetic moments of 6.6, 5.6, and 3.6 μ_B, respectively, with orbital moments of about 3, 3, and 2 μ_B, respectively, as confirmed by applying the XMCD sum rules [10,11]. The magnetic moment per atom and related dichroism decrease dramatically as the impurity form clusters. More data collected during the experiment are being currently analyzed.

In conclusion, Fe, Co, and Ni transition metal impurities on K films display localized atomic configurations with fully unquenched orbital magnetic moments. The present results conclusively prove that the giant moments in alkali systems originate from the localization of the 3d states. Finally, we demonstrated that combined XAS-XMCD experiment have a yet unexplored potential for the study of diluted systems, giving simultaneous and direct access to the electronic and magnetic configuration of the impurity states.

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FIGURE 1: XAS spectra over the $L_{2,3}$ edges recorded with parallel (solid line) and antiparallel (dotted line) to the applied magnetic field for (a) 0.015 ML Fe, (b) 0.015 ML Co, and (c) 0.004 ML Ni deposited on K films. In each case the resulting XMCD is also shown.

FIGURE 2: (a) XAS and XMCD spectra over the $L_{2,3}$ edges recorded with parallel (solid line) and antiparallel (dotted line) to the applied magnetic field for a thick Co film (30 ML). (b) Comparison of the XMCD for Co impurities on a K film and 30 ML Co.

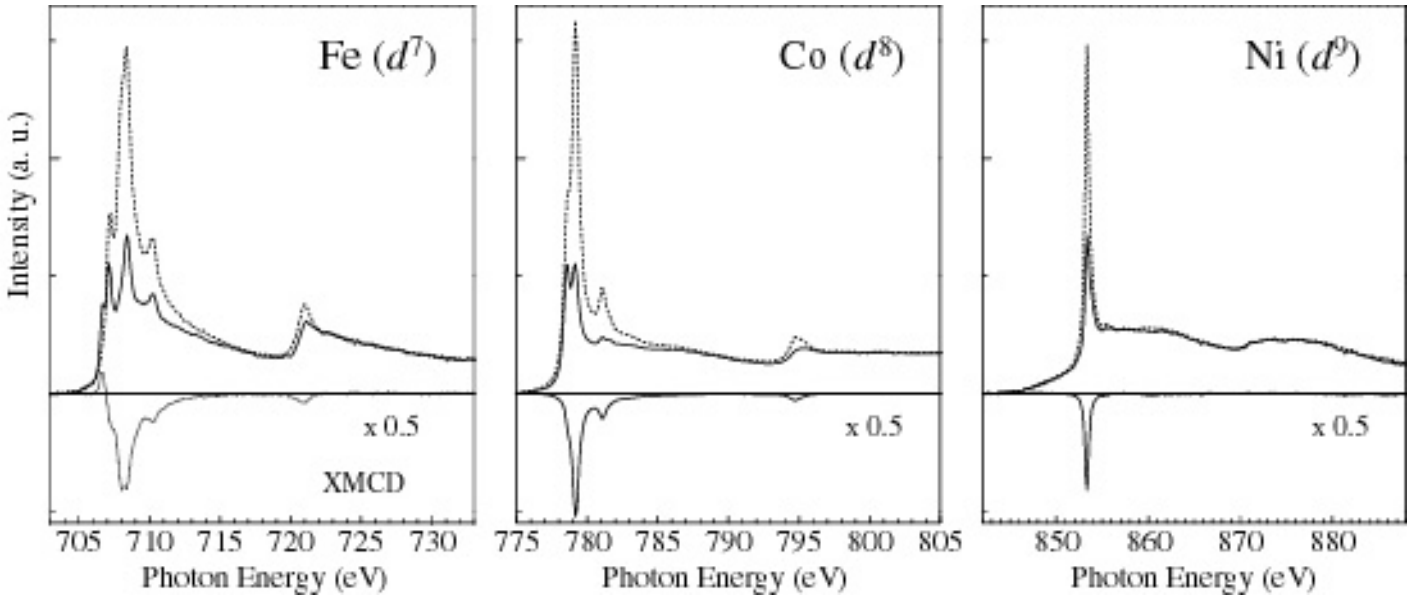

Fig. 1: HE 991 report

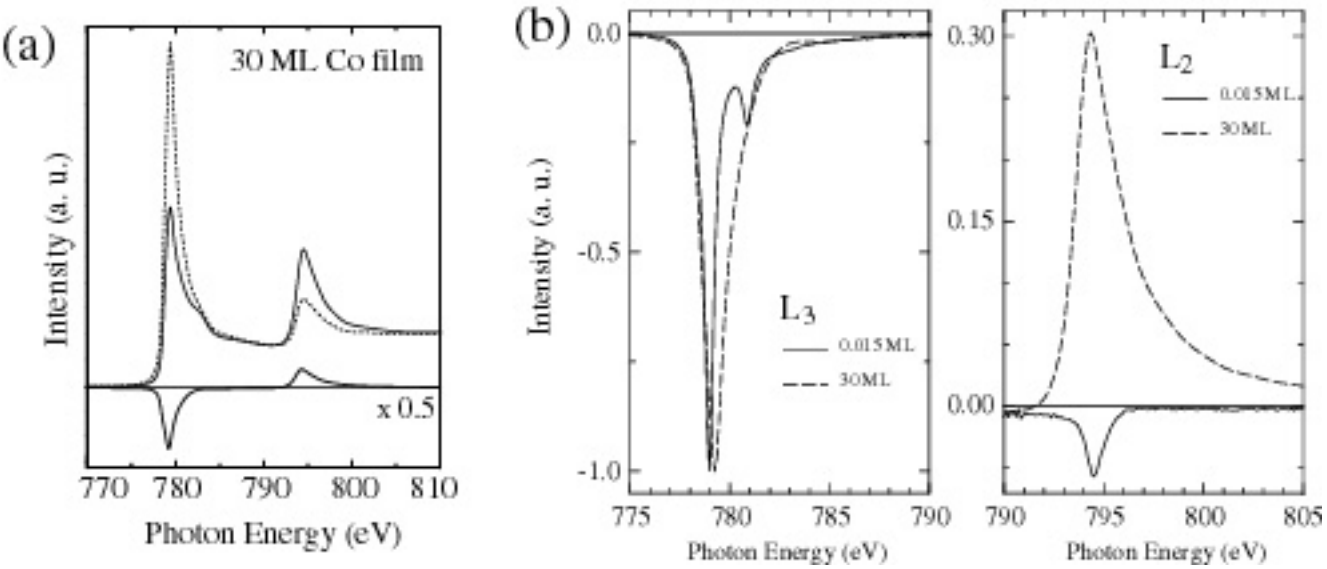

Fig. 2: HE 991 report