ESRF	**Experiment title:** Magnetic moments and anisotropy of transition metal impurities on simple metal films	**Experiment number**: HE 1221
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Report:

Most transition metal atoms possess large spin and orbital magnetic moments. When such atoms are dispersed as impurities in a free-electron medium as, e.g., noble or simple metals, however, the magnetic moment may vary greatly or even disappear depending on the atomic *d*-states valence, host electron density, and on the temperature [1]. Charge transfer and/or spin flip scattering mechanisms such as spin fluctuations and Kondo screening are thought to be responsible for such effects. Experiment HE 1221 aimed at characterizing the electronic structure and related magnetic behavior of Fe and Ni impurities on simple metal hosts with different free-electron densities ρ. In this respect, metals in the alkali series and Cu represent ideal hosts (with ρ ranging from 1.4 to 8.5 x 10^{22} electrons / cm^3, Ref. 2) to study the extent to which the transition metal *d*-states interact with conduction electrons. Previous experiments showed that the magnetic moment of Ni atoms does not survive in Na and Li hosts [3,4], while it is ionic-like in K [3,5]. No explanation could be given for this effect since only the total magnetization of the impurities was measured. Our approach is to combine x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) to gather information on the valence character of the impurities and, at the same time, measuring their local spin and orbital magnetic moments. We show that the disappearence of the Ni magnetic moment on Na is due to increased charge transfer from the conduction bands into the Ni *d*-states, which exhibit a mixed d^9-d^{10} character. In contrast, the Fe moment survives on Na and, although strongly reduced, also on the higher ρ substrates Li and Cu. The Fe *d*-states are shown to be ionic-like on K and to hybridize progressively with increasing ρ. The XAS-normalized XMCD signal of Fe on Cu(100) at 10 K is weaker in the single adatom limit compared to 1 monolayer Fe, suggesting that Kondo screening might reduce the magnetic response of the impurities.

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, and Ni were evaporated in small quantities (0.01 – 0.02 ML) on Cu(100) as well as on K, Na, and Li films predeposited on Cu(100). XAS at the $L_{2,3}$ edges was performed in total electron yield mode using circularly polarized light in a 7 T magnetic field with the sample at T=10 K. XMCD was recorded by switching both the light polarization and the sample magnetization.

Figure 1(a) shows the XAS spectra recorded for Ni impurities deposited on a Na film for parallel (solid lines) and antiparallel (dotted lines) alignment of the light polarization with respect to the applied field. The XMCD spectra is also shown at the bottom of the panel. Contrary to Ni/K, Ni/Na impurities do not present any XMCD, implying a nonmagnetic ground state. Density functional and jellium calculations predict this effect based on the broadening of the impurity virtual bound state associated with the increased electron density of Na hosts compared to heavier alkali elements [6]. By comparing the isotropic XAS spectra for 0.004 ML of Ni on Na and on K in Fig. 1(b), we argue that the Ni states in Na assume a significant d^{10} character, and that this has a direct bearing on the quenching of the Ni moment. The intensity of the L_3 edge due to the $3d^9 \rightarrow 2p^5 3d^{10}$ transition is reduced by more than 50 % for Ni/Na compared to Ni/K. The width of the line, however, although larger compared to Ni/K (0.80 vs 0.52 eV FWHM) is still much narrower relative to bulk Ni spectra (~1.7 eV, Ref. 7), indicating that Ni valence states can still be considered as predominantly localized. These observations suggest the presence of two resonant d^9 and d^{10} configurations of ionic type [8], where fast incoherent electron hopping (i.e. charge fluctuation) forbids the existence of a magnetic moment.

Owing to its almost half empty *d*-shell, Fe does not loose completely its magnetic moment as the substrate ρ increases moving up in the alkali series and finally to Cu. Figures 2(a)-(d) show the XAS and XMCD spectra of Fe impurities on K, Na, Li, and Cu. The magnitude of the XMCD signal measured at 7 T reveals that the local Fe moment is strongly reduced on Li and Cu compared to K and Na. The sign of the L2 XMCD reverses in going from K to Cu, indicating a significative decrease of the orbital moment relative to the spin moment [9]. Parallely, the XAS multiplet structure observed on K and Na broadens and eventually disappears on Cu. This trend is a clear indication of the increased hybridization of the Fe *d*-states with the substrate valence band. The substrate ρ turns out to be the crucial parameter determining the extent to which the Fe *d*-states mix with the host conduction electrons and thereby governing the magnitude of the impurity magnetic moment.

The XAS-normalized XMCD signal of Fe adatoms on Cu(100) at 10 K is weaker compared to 1 monolayer Fe, contrary to what happens for Fe on K. Since diluted Fe:Cu alloys constitute a well-known Kondo system with Kondo temperature T_K = 20-30 K in the bulk [1], it is reasonable to ask whether the reduced magnetization can be ascribed to partial Kondo screening of the Fe moment by the Cu conduction electrons at 10 K. To answer this question, the temperature dependence of the XMCD will be addressed in the future for Fe and other impurities (Co) on Cu(100) that present T_K in an accessible and convenient range.

[1] *Magnetic Properties of Metallic Alloys*, edited by H. Suhl (Academic Press, New York, 1973).
[2] N.W, Ashcroft and N.D. Mermin, *Solid State Physics*, (Saunders, Philadelphia, 1976).
[3] R. Kowallik et al., *Phys. Rev. Lett.* **63**, 434 (1989).
[4] G. Bergmann and M. Hossain, *Phys. Rev. Lett*. **86**, 2138 (2000).
[5] P. Gambardella, S.S. Dhesi, S. Gardonio, C. Grazioli, P. Ohresser, and C. Carbone, *Phys. Rev. Lett.* **88**, 047202 (2002).
[6] N. Papanikolau, N. Stefanou, R. Zeller, and P.H. Dederichs, *Phys. Rev. B* **46**, 10 858 (1992).
[7] C.T. Chen et al., *Phys. Rev. B* **42**, 7262 (1990); S.S.Dhesi et al., *ibid*. **60**, 12852 (1999).
[8] L.L. Hirst, *Phys. Kondens. Mater.* **11**, 255 (1974).
[9] B.T. Thole, P. Carra, F. Sette, and G. van der Laan, *Phys. Rev. Lett.* **68**, 1943 (1992); P. Carra, B.T. Thole, M. Altarelli, and X. Wang, *ibid*. **70**, 694 (1993).

FIGURE 1: (a) XAS spectra over the $L_{2,3}$ edges recorded with parallel (solid line) and antiparallel (dotted line) to the applied magnetic field for 0.004 ML Ni deposited on a Na film. The resulting XMCD is also shown. (b) Comparison between the $(I_+ + I_-)$ spectra, normalized to the *s* edge jump, for 0.004 ML Ni on Na (solid line) and on K (dashed line).

FIGURE 2: XAS and XMCD spectra over the $L_{2,3}$ edges recorded with parallel (black line) and antiparallel (red line) to the applied magnetic field for Fe impurities (0.02 ML) on K (a), Na (b), Li (c), and Cu (d).

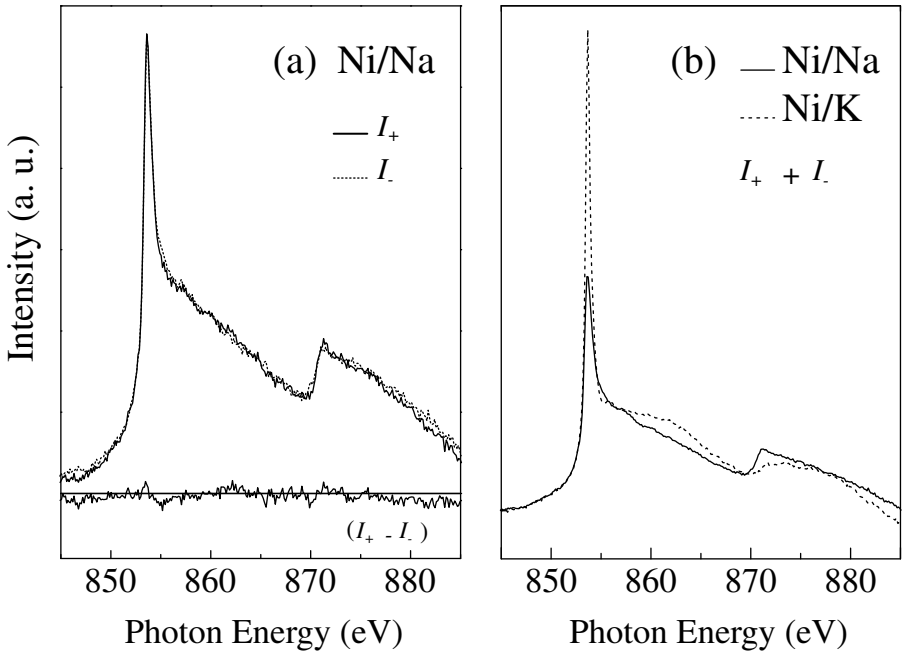

Fig. 1: HE 1221 Report

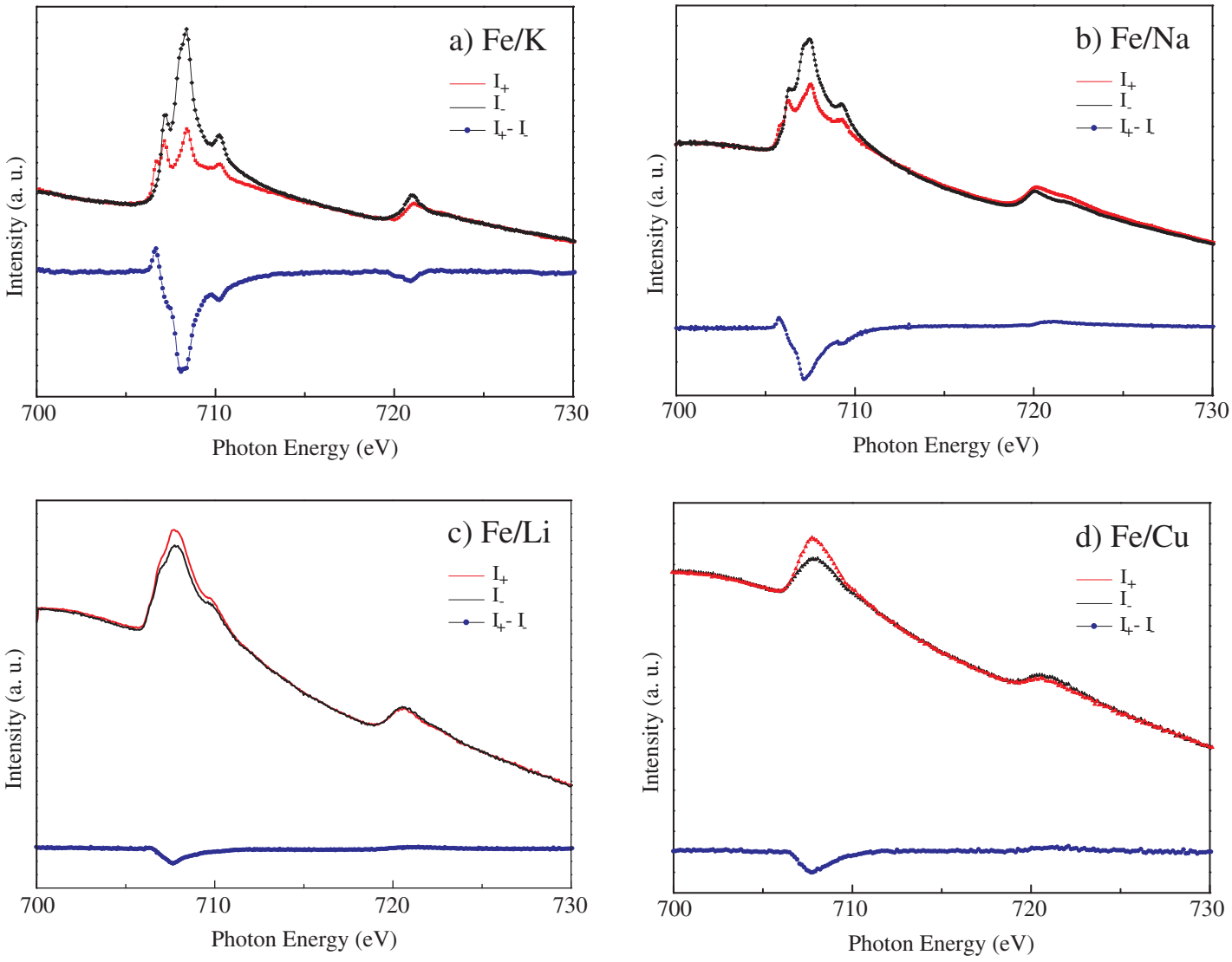

Fig. 2: HE 1221 Report