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LETTER TO THE EDITOR

Magnetic circular dichroism in Ni 2*p* photoemission of ferromagnetic nickel

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Abstract. The Ni 2*p* photoemission of nickel metal measured with circularly polarized synchrotron radiation is compared with results of spin-polarized relativistic Korringa–Kohn–Rostoker Green's function calculations. The magnetic circular dichroism displays pronounced satellite structures over a wide energy range which are not reproduced by the one-particle calculation. This gives a clear indication for the presence of electron-correlation effects.

1. Introduction

In the last decade, much insight into the magnetic and electronic properties of 3*d* transition metal systems has been gained from magnetic circular dichroism in x-ray absorption (MCD-XA), especially by applying the sum rules [1] and by using first-principles band-structure calculations [2] to analyse the data. Ni metal served as a paradigm in the prediction of magneto-optical effects in the soft x-ray range [3], and although already a quarter of a century old, the interpretation is still controversial for this material. Both itinerant and localized models provide an incomplete description for the high-energy spectroscopy of nickel metal [4]. For instance, the Ni 2*p* and 3*p* XA spectra show distinct satellite structure [5,6]. Wu *et al* [7] ascribed the 6 eV satellite in the Ni 2*p* XA to a van Hove singularity at the *L* critical point in reciprocal space. The band structure at this point contains a large amount of $d(xz, yz)$-like wavefunction components, which do not contribute to the MCD. Therefore, the presence of a 4 eV satellite observed in MCD [8] remains puzzling in a one-particle model. Anderson impurity calculations, on the other hand, provide a natural explanation of the satellite structure [5], but do not take the band dispersion into account. Nevertheless, satellite features due to electron correlation are often negligibly small in XA because the core hole is efficiently screened by the electron excited into the empty *d* state.

In this Letter we direct our attention to the photoemission (PE). Contrary to XA, the core hole in PE is not screened by the excited electron. Therefore, many-body effects can induce strong satellite structures, which are absent in a one-particle model where Coulomb and exchange interaction and electron correlation are taken into account by an effective potential. We will use MCD to study these correlation effects [9].

The valence electrons affect the core-level spectrum due to screening of the core hole in the final state. In free-electron metals the binding energy (BE) of the core state will be lowered

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by collective screening from the itinerant valence electrons. In localized materials we expect, apart from a well screened core hole state at low BE, also poorly screened states at higher BE. This effect is well known in the gas phase but is usually less pronounced in metals where many different screening channels lead to an overall broadening and asymmetry of the PE lines. The one-particle model is a useful tool for identifying multi-electron excitations. The latter will not show up in these calculations, whereas most other effects, such as multiple scattering, are included.

MCD-PE, first performed at the Fe $2p$ core level of ferromagnetic iron [10], has lent itself to a qualitative explanation [11, 12]. The $2p$ PE of $3d$ transition metals is thought to be especially suited for a simple analysis because the core spin–orbit interaction splits the spectrum into two well separated structures [13]. Fully relativistic calculations indicate that the exchange field on the core level is of the order of tenths of eV, which leads to almost equidistantly spaced magnetic sublevels within the two j levels [11, 14, 15]. There has been much interest in extracting the magnitude of the effective exchange field from the experimental Fe $2p$ MCD data using the six magnetic sublevels which emerge from a single-particle model [14, 16–19]. However, high-resolution spectra of Fe metal revealed that the MCD signal is strong everywhere between the $2p_{3/2}$ and $2p_{1/2}$ levels [20]. This disallows the simple approach of simulating the core level PE by six magnetic sublevels [21].

In localized systems such as rare-earths, many-body effects lead to pronounced multiplet structure due to Coulomb and exchange interactions [22–24]. Nickel metal is considered to be at the borderline of itinerant and localized behaviour and is therefore an interesting test case to study.

2. Experimental details

Ultra-thin Ni films of several monolayers (ML) thick were prepared by deposition onto 4 ML Co films epitaxially grown on Cu(100). The Ni and Co films grow with in-plane face-centred cubic (fcc) periodicity as verified by high-resolution low energy electron diffraction (HRLEED), which is described elsewhere [8]. Photoemission measurements on Ni films of various thicknesses were done using 85% circularly polarized light from beamline ID12B of the European Synchrotron Radiation Facility (ESRF) at Grenoble. We used a photon energy of 1100 eV, so that the Ni $2p$ MCD signal was relatively large [25]. A current pulse through a nearby coil was used to give remanent magnetization in the surface plane. The photoelectrons were collected at normal emission with the x-rays incident at 60° from the surface normal in the plane spanned by the sample magnetization and light helicity. Use of a hemispherical multichannel analyser with an acceptance angle of 40° ensured an averaging over intensity variations due to multiple scattering effects.

3. Computational method

The calculations presented in this paper were performed using the fully relativistic spin-polarized relativistic Korringa–Kohn–Rostoker (SPR-KKR) Green's function method [26]. Based on a one-particle scheme the corresponding core level PE spectra were calculated using the formalism derived by Ebert *et al* [27, 28] where spin–orbit coupling and spin polarization are treated on equal footing. We considered bulk-like fcc-Ni with a lattice parameter of 6.652 au. The spectra were calculated for normal incidence of the photons and using an angle integrated mode. The final states were represented by time reversed LEED (low electron energy diffraction) states. Because of the rather high photon energy (1100 eV) a relatively

high l-expansion was used for the final states ($l_{max} = 7$). To deal with multiple scattering of the electrons in the final states the real-space KKR method was used. The influence of multiple scattering in the $2p$ spectrum is strongly reduced by the angle integration over the final states due to the large acceptance of the PE detector. Multiple scattering effects in the calculated spectra were only visible at narrow acceptance angles and low kinetic energies. The calculated spectra were convoluted to account for intrinsic and apparative broadening using a Lorentzian of $\Gamma = 0.5$ (1.3) eV for the $2p_{3/2}$ ($2p_{1/2}$) region and a Gaussian of $\sigma = 0.15$ eV, respectively. The calculated relative BE of the magnetic sublevels is given in table 1.

Table 1. Calculated binding energies (eV) of the Ni $2p$ magnetic sublevels in nickel metal. The energy is taken relative to the average over the $2p_{3/2}$ level.

BE (eV)	j, m_j
−0.157	3/2, −3/2
−0.052	3/2, −1/2
0.052	3/2, +1/2
0.155	3/2, +1/2
17.395	1/2, +1/2
17.496	1/2, −1/2

4. Discussion

Figure 1 shows the Ni $2p$ photoemission spectrum averaged over the two light helicities together with the MCD signal. For comparison, the sign of the MCD-PE was chosen as in [29, 30].

At high kinetic energies the $2p$ core electron is excited into an empty continuum state which has no interaction with the spin-polarized d states. Dichroism in PE, therefore, does not arise due to the Pauli principle, such as in XA, but can be ascribed to the exchange interaction between the spin polarized valence states and the spin–orbit split core state. The dichroism signal is proportional to the orbital polarization in the magnetic sublevels of the core hole [11, 29]. The combined effect of exchange and core spin–orbit interaction leads to a shape of the MCD that is (+, −) for the $j = 3/2$ and (−, +) for the $j = 1/2$ core level. Such a shape is clearly confirmed by the calculated spectrum shown in figure 1.

The experimental $2p_{3/2}$ structure shows a well screened peak at the threshold of excitation followed by a poorly screened peak at 6 eV higher BE. If the energy separation between these peaks is large, the MCD signal of both should have a (+, −) shape. There is, however, a considerable hybridization between both states resulting in a transfer of spectral intensity between main and satellite peak. It is also seen that the poorly screened peak is much broader than the well screened peak, which indicates remnant atomic multiplet structure. The next pronounced feature in the experimental MCD is a negative peak around ∼16 eV, which is just below the threshold of $2p_{1/2}$ excitation. This peak must therefore be due to a poorly screened $2p_{3/2}$ final state (a similar feature is replicated at ∼16 eV above the $2p_{1/2}$). The energy overlap of the two spin–orbit split regions complicates the analysis of the spectral structure at higher BE. A further analysis using a many-body calculation, such as given in [30], is required. The shape of the experimental spectrum agrees quite well with the prediction from this calculation, albeit that the relative satellite intensities are somewhat different. A more detailed comparison of the Ni $2p$ spectrum with a final state impurity model will be presented elsewhere [31].

Although the size of the MCD depends on the details of the energy broadening, it is clear from figure 1 that the measured magnitude roughly agrees with the theoretical expectation. Thus, the MCD of nickel is about 5× smaller than that of iron [20]. The measured MCD of

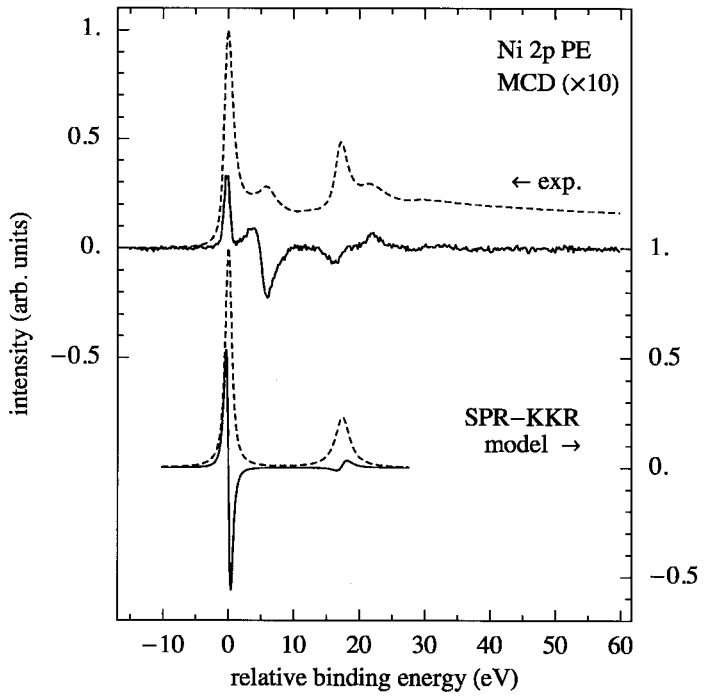

Figure 1. Experimental and calculated Ni $2p$ photoemission of nickel metal at a photon energy of 1100 eV using wide-angle photoelectron detection. The MCD spectra (dashed lines) are given on a 10× enhanced intensity scale with respect to the helicity averaged spectra (drawn lines). The calculations, performed using the spin-polarized relativistic Korringa–Kohn–Rostoker Green's function method, include multiple scattering but exclude electron correlation effects.

the Ni $2p_{1/2}$ final states is larger than expected from the one-particle calculation, cf figure 1, which can be ascribed to the strong mixing with the $2p_{3/2}$ final states through core–valence interactions.

5. Conclusions

In correlated materials, such as nickel, the Coulomb energy depends on the local d-state occupation, where the PE spectrum shows well screened and poorly screened peaks with an energy separation which is mainly determined by the on-site dd Coulomb interaction. We found that the MCD in PE gives clear evidence that the features observed at higher BE are due to electron correlation effects. Using the SPR-KKR method to calculate the MCD we were able to distinguish between one-particle and many-body effects.

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