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First principles relativistic theory of photoemission from magnetic surfaces

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Abstract

A first principles theory of spin- and angle-resolved photoemission has been developed. The theory is based on density functional theory and is fully relativistic. It is implemented using multiple scattering theory with a Green's function calculated using a real space cluster method. No lattice symmetry or periodicity is assumed and therefore our approach can be applied to low-dimensional systems. We illustrate the theory with a calculation of the photoemission spectra from Ni(1 0 0) and interpret the result in terms of the band structure. The effect of the polarisation of the photon on the spectra is emphasised.

Keywords: Photoemission; Band structure; 3d metals

A one-electron picture of photoemission can be described by an application of the Golden rule between initial state i and final state f. The Golden rule may be written as [1]

$$T = -\frac{2}{\hbar}\sum_f \iint \psi_f^\dagger(\mathbf{r})\hat{H}'(\mathbf{r})\,\mathrm{Im}\,\mathbf{G}^r(\mathbf{r},\mathbf{r}';\varepsilon_i) \times \hat{H}'^\dagger(\mathbf{r}')\psi_f(\mathbf{r}')\mathrm{d}^3\mathbf{r}\;\mathrm{d}^3\mathbf{r}', \quad (1)$$

where

$$H'(\mathbf{r}) = -ec\alpha\cdot\mathbf{A}(\mathbf{r}) \quad (2)$$

is the perturbation responsible for the transition and contains the electron–photon interaction. $\mathbf{G}^r(\mathbf{r},\mathbf{r}';\varepsilon_i)$ is the retarded Green's function at energy ε, and ψ_f is the time reversed LEED state describing the ejected electron. The Green's function is written in the standard τ–matrix representation in terms of the regular and irregular single-particle solutions of the Kohn–Sham–Dirac equation of relativistic density functional theory and the matrix elements of the scattering path operator [1]. The scattering potential $V(\mathbf{r})$ consists of an array of non-overlapping muffin-tin potentials representing the lattice of atoms. Outside the muffin-tins a constant potential is assumed.

We have obtained a fully relativistic photocurrent formula which is valid for any atomic arrangement and is a relativistic generalisation of the theory of Ernst et al. [2].

Our approach for evaluating Eq. (1) is the real-space cluster method. This distinguishes our work from earlier implementations which were always k-space based. In this case the τ-matrix can be obtained by inversion of the KKR matrix: The real-space KKR method can be applied to any arrangement of scatterers.

We illustrate our theory with an application to Ni [3] where the measured exchange splitting is half of the magnitude of the calculated splitting and the d-band width is approximately 30% smaller than the calculated width [4]. Spin-resolved photoemission experiments have determined exchange splittings Δ_{ex} in Ni and have shown that it depends strongly on the energy, wavevector and the symmetry of the band [5]. We have performed calculations with photon energies of 11 and 44 eV at

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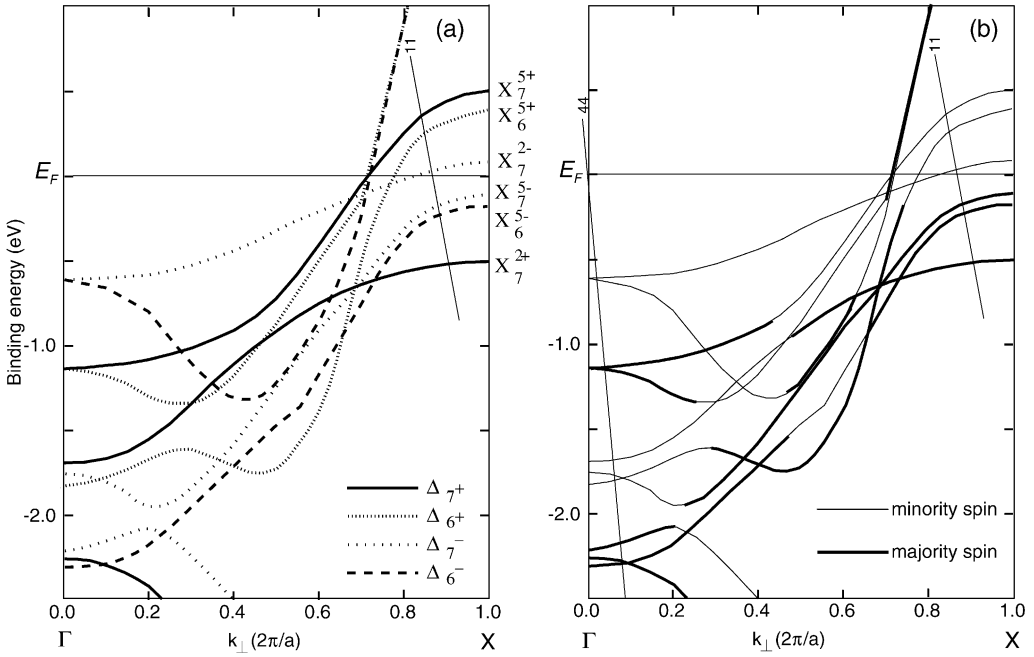

Fig. 1. Band structure of Ni(0 0 1). (a) Bands exhibiting the double group symmetry. Thin solid lines are sections of the final-state band, shifted down by the photon energy indicated. (b) As in (a), but with the bands distinguished by their minority (majority) spin expectation value as thin (thick) lines.

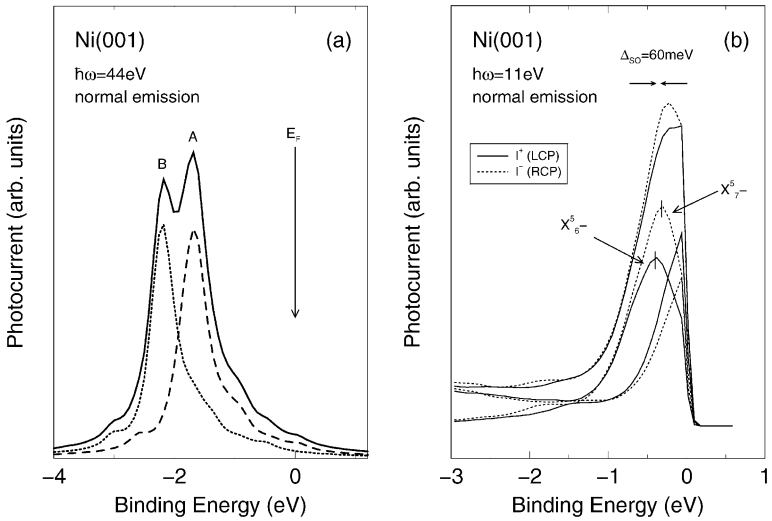

Fig. 2. Spin resolved photoemission spectra for two photon energies (a) Normal emission spectra for right-circularly-polarized photons of 44 eV incident normal to the Ni(0 0 1) surface. The solid line is total emission, majority (minority) spectra are dotted (dashed) lines. (b) Spin-orbit splitting at the X-point.

binding energies close to the Fermi level to gain insight into the details of the d-band structure of Ni(0 0 1). The calculation of the photocurrent is performed with broadening in the energy of 0.01 Ry and processed with a Fermi–Dirac distribution at room temperature for close comparison with experiment.

In Fig. 1(a) the theoretical band structure along the Δ-axis for the (0 0 1) surface is presented. The bands are displayed with different symbols according to their symmetry. Classifications according to spin-occupancy are shown in Fig. 1(b). Thick (thin) lines classify the bands according to whether they are majority (minority) spin.

The complexity of the Ni band structure can be attributed to the interplay between the exchange and spin-orbit interactions which are of the same order of magnitude ($\approx 0.3/0.1$ eV).

Fig. 2 shows spin-resolved normal photoemission spectra of Ni(0 0 1) obtained with incident radiation at energies of 44 and 11 eV, probing states close to Γ and $k_{\perp}(2\pi/a) = 0.8$ respectively. Fig. 2(a) shows that, close to Γ, the spin-resolved spectrum displays two distinct peaks, labeled A and B, at -1.6 and -2.2 eV binding energies respectively. Due to the experimental geometry, only initial states with Δ_5 spatial symmetry are observed. Comparison with the valence-band structure identifies these peaks as direct transitions from the Δ_7^5+ valence band with predominantly minority spin characteristics and the Δ_6^5- valence band with predominant majority spin. The equivalent labels in Fig. 2(a) mark where the initial state band crosses the Δ_1 final state band shifted down by 44 eV. This figure clearly gives a value for the exchange splitting at Γ of 0.6 eV.

Dichroism asymmetry makes it possible to determine information about important parameters of the relativistic band structure. At 11 eV photon energy for example, two majority spin bands of Δ_7- and Δ_6+ contribute the bulk of the signal. They are energetically separated by the spin-orbit interaction, and appear each in different helicity spectra. The separation of the corresponding peaks in the right- (RCP) and left-circularly-polarized (LCP) spectra of Fig. 2(b) is then a measure of the strength of spin-orbit coupling in the valence bands. At this particular point of the Brillouin zone, 60 meV is obtained, which compares very well with the separation of X_6^5- and X_7^5- critical-points in Fig. 2(a).

In this paper we have reported on the development of a real-space fully-relativistic spin- and angle-resolved photoemission code in the independent particle approximation. We have shown that it yields good agreement with experiment and may be used to give unique insight into the details of the band structure of magnetic materials.

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