



Spin-polarization in ccv Auger electron spectroscopy for transition metals

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Abstract

The contributions of core states and the valence band to spin-resolved ccv Auger electron spectra (AES) are discussed in terms of a relativistic, spin-polarized theory. The influence of the core-state spin polarization and of selecting particular valence bands via cross sections is shown for the surface layer of BCC Fe(1 0 0) as calculated by means of the Screened KKR ab-initio electronic structure method. © 2002 Elsevier Science B.V. All rights reserved.

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In a relativistic multiple scattering theory [1–3], Auger electron spectroscopy (AES) can be formulated in a spin-polarized manner that permits simultaneously to calculate transition matrix elements and the DOS. A similar approach was presented for the core–valence–valence (cvv) case [4]; however, if one is interested in the valence band properties it seems to be more convenient to analyze two core states, i.e., ccv AES, instead of convoluting the valence bands.

For a ccv AES process, we have to describe the electronic transitions from initial states c_2 —the upper core states and v —the valence states, and from the final states c_1 —the lower core states and A —the outgoing ‘Auger’ electron. Obviously, there are two possible transition processes: the direct (D) process $c_2 \rightarrow c_1$ and $v \rightarrow A$ and the exchange (E) process $c_2 \rightarrow A$ and $v \rightarrow c_1$. Thus, the emission intensity as given by

$$I(\varepsilon) = \delta(\varepsilon_2 - \varepsilon) \overline{|D - E|^2} = I^{c_1 c_2}(\varepsilon; s, \hat{p}), \quad (1)$$

can be determined as spin (s) and angle (\hat{p}) resolved. Inserting the actual involved states and the electronic interaction as a static Coulomb interaction into the

direct and exchange integrals the transition intensity can, as usual, be decomposed into a matrix of cross sections $\sigma_{Q_v}^{c_1 c_2}(\varepsilon; s, \hat{p})$ and the DOS defined in multiple scattering theory by the imaginary part of the scattering path operator $\text{Im}\tau_{Q_v}(\varepsilon)$

$$I^{c_1 c_2}(\varepsilon; s, \hat{p}) = -\frac{1}{\pi} \sum_{Q_v} \sigma_{Q_v}^{c_1 c_2}(\varepsilon; s, \hat{p}) \text{Im}\tau_{Q_v}(\varepsilon), \quad (2)$$

that includes a sum over all valence states $Q_v = (\kappa\mu)$.

In a spin-polarized relativistic treatment, all involved states are sums of the coupled solutions (Q') of the following form

$$\psi_{Q'}(\mathbf{r}) = \sum_{Q''} \begin{pmatrix} g_{Q'' Q'}(r) & \chi_{Q''}(\hat{r}) \\ i f_{Q'' Q'}(r) & \chi_{Q''}(\hat{r}) \end{pmatrix}. \quad (3)$$

In the case of core states, $g_{Q'' Q'}(r)$ and $f_{Q'' Q'}(r)$ are the solutions of the coupled Dirac equation for a given core eigenvalue ε (see Ref. [2]), the $\chi_{Q''}(\hat{r})$ being spin-spherical harmonics. The valence states are described by regular scattering solutions and the DOS is given as discussed in Ref. [2]. Continuum states at energies considerably high above the Fermi level can be described in the single-site (final state) approximation.

The matrix of cross sections as introduced in Eq. (2) $\sigma_{Q_v}^{c_1 c_2}(\varepsilon; s, \hat{p})$ can be separated into three terms according

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to the three different transition processes

$$\sigma_{Q_v}^{c_1 c_2} = \sigma_{Q_v}^{c_1 c_2, D^2} + \sigma_{Q_v}^{c_1 c_2, E^2} + \sigma_{Q_v}^{c_1 c_2, DE}. \quad (4)$$

All of them show the same formal structure, namely,

$$\begin{aligned} \sigma_{Q_v}^{c_1 c_2, D^2}(\varepsilon; s, \hat{p}) = & \sum_{Q_A Q'_A} i'^{\lambda - l_A} C(l_A \ 1/2 j_A; \mu_A - s, s) \\ & \times C(l'_A \ 1/2 j'_A; \mu'_A - s, s) \\ & \times Y_{l_A, \mu_A - s}(\hat{p}) Y_{l'_A, \mu'_A - s}^*(\hat{p}) \\ & \times \mathbf{I}(Q_{c_1} Q_{c_2} | Q_A Q_v) \\ & \times \mathbf{I}(Q_{c_1} Q_{c_2} | Q'_A Q_v), \end{aligned} \quad (5)$$

i.e., it contains sums over the continuum states Q_A and Q'_A , as well as integrals \mathbf{I} corresponding to different combinations of initial and final states

$$\sigma_{Q_v}^{c_1 c_2, D^2} \Rightarrow \mathbf{I}(Q_{c_1} Q_{c_2} | Q_A Q_v) \mathbf{I}(Q_{c_1} Q_{c_2} | Q'_A Q_v),$$

$$\sigma_{Q_v}^{c_1 c_2, E^2} \Rightarrow \mathbf{I}(Q_{c_1} Q_v | Q_A Q_{c_2}) \mathbf{I}(Q_{c_1} Q_v | Q'_A Q_{c_2}),$$

$$\begin{aligned} \sigma_{Q_v}^{c_1 c_2, DE} \Rightarrow & \mathbf{I}(Q_{c_1} Q_v | Q_A Q_{c_2}) \mathbf{I}(Q_{c_1} Q_{c_2} | Q'_A Q_v), \\ & + \mathbf{I}(Q_{c_1} Q_{c_2} | Q_A Q_v) \mathbf{I}(Q_{c_1} Q_v | Q'_A Q_{c_2}). \end{aligned} \quad (6)$$

The D^2 term results in an Auger electron spin polarization due to the valence band while the E^2 term produces a spin polarization due to the c_2 core states. Thus, besides the value of the valence-band spin polarization the resulting Auger electron spin polarization also depends on the spin character of the core states weighted by the amplitudes of the radial parts in the different terms. Only for vanishing E^2 and DE terms the valence-band spin is transferred to that of the Auger electron.

In the present contribution, we shall investigate AES integrated over the angle of the emitted electron \hat{p} , for which the corresponding cross sections can easily be derived from Eqs. (5) and (6). An experimentally observed ccv Auger electron spectrum results from all different atoms emitting electrons and from all possible core state combinations for one particular atom corresponding in energy

$$I^{\text{exp}}(\varepsilon; s) = \sum_{\text{atoms}} \sum_{c_1, c_2} I^{c_1 c_2}(\varepsilon; s). \quad (7)$$

Note that, the energy scale of each $I^{c_1 c_2}(\varepsilon; s)$ has to be aligned considering the different energy eigenvalues of each core state. We found the largest core state split in the Fe surface atom of 1.1 eV for the L_3 ($2p_{3/2}$) states and of 2.5 eV for the M_3 ($3p_{3/2}$) states. The $L_3 M_{23} V$ AES consists of 24 separate processes involving the four L_3 and the six M_{23} states.

The electronic structure was calculated for the system BCC Fe(100)/Fe₆/Vac using the Screened KKR method. Here, only the results for the surface Fe layer are shown. In Fig. 1, we show the total $L_3 M_{23} V$ AES for the Fe surface layer. Almost 70% of the intensity is due to

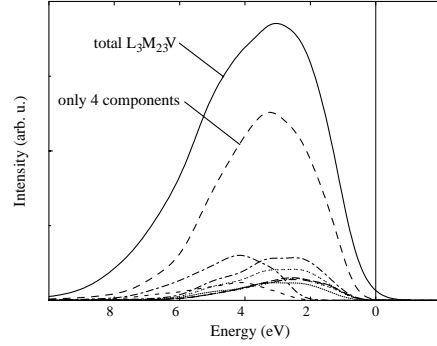


Fig. 1. Total (spin up + spin-down) $L_3 M_{23} V$ AES of the surface layer of BCC Fe(100) (full line). Dashed curve is the sum of the four largest total partial AES. All other curves are the largest spin-resolved partial AES.

transitions from only four of the 24 core-state combinations, namely, the ones with $\Delta\kappa_c = 0$ ($\Delta j_c = 0$) and $\Delta\mu_c = 0$ (dashed curve). For $Q_{c_1} = L_3$ and $Q_{c_2} = M_{23}$ these are the core states with $j_c = 3/2$ and (i) $\mu_c = -3/2$, (ii) $\mu_c = -1/2$, (iii) $\mu_c = +1/2$, and (iv) $\mu_c = +3/2$. The 8 partial spin-resolved spectra for these four cases are also shown in Fig. 1. We find most of the intensity for cases (i) and (iv) corresponding to respective opposite spins. It is interesting to mention that (i) and (iv) are the only uncoupled states and they show a spin polarization of 100% with a different sign.

In the following we compare the results for cases (i) and (iv). In Figs. 2 and 3 the spin-up and spin-down spectra and the cross sections are shown together with the corresponding DOS for the valence band. First of all it is important to note that the DOS in both figures is displayed for selected (κ, μ) valence states and, consequently, is not spin-polarized. Figs. 2(b) and 3(b) exhibit the largest cross sections for both spin directions for cases (i) and (iv) of the outgoing electron, respectively. The resulting unbroadened spectra are shown in (c) for case (i) and in (d) for case (iv). We find large amplitudes for opposite spins, see Figs. 2(d) and 3(c), as already pointed out.

For all spectra the respective largest partial valence-band state resolved spectra are shown. The difference in the form of the spin-up and the spin-down spectra is given by the contribution of different valence-band states, see in particular the peak around 2 eV in Fig. 3(c), which occurs only in the spin-down spectrum. Furthermore, in the spin-up case there is no dominating partial spectrum: almost all d-band partial spectra yield a comparable contribution. In the spin-down case we find in both cases (i) and (iv) a dominating contribution from the uncoupled ($j = 5/2$, $\mu = +5/2$) valence state and a comparably big one from only two other states.

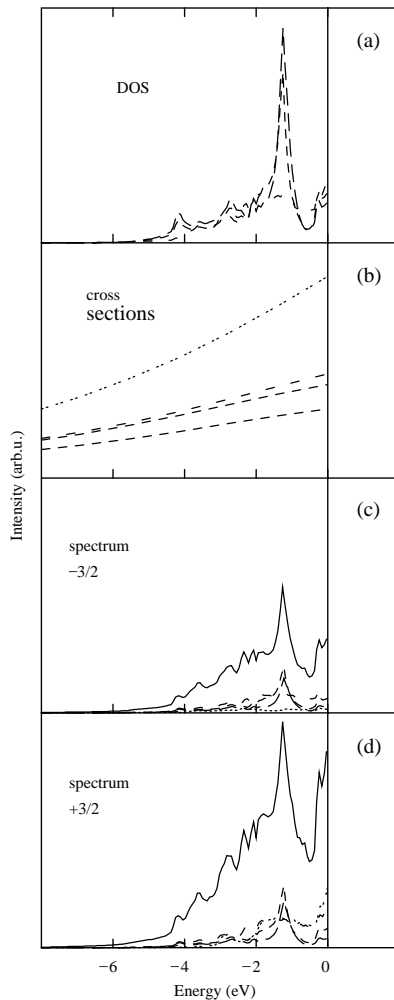


Fig. 2. Spin-up partial $L_3M_{23}V$ AES for (c) $Q_{c_1} = Q_{c_2} = (\kappa = -2, \mu = -3/2)$ and (d) $Q_{c_1} = Q_{c_2} = (\kappa = -2, \mu = +3/2)$ together with the largest valence-band state resolved contributions to the spectrum in (c) and (d), respectively, and the corresponding DOS in (a) and cross sections in (b).

Summarizing, in the case of core transitions $\Delta j_c = 0$ and $\Delta \mu_c = 0$, we find remarkable contributions to the AES from (selected or all) d-band valence states and that the spin-resolution of the spectrum is mainly determined by the spin character of the core-states. Calculations of other AES (for $\Delta j_c \neq 0$ and/or $\Delta \mu_c \neq 0$) show couplings to s-like or p-like valence states for which the cross sections can have a very different energy dependency and amplitudes.

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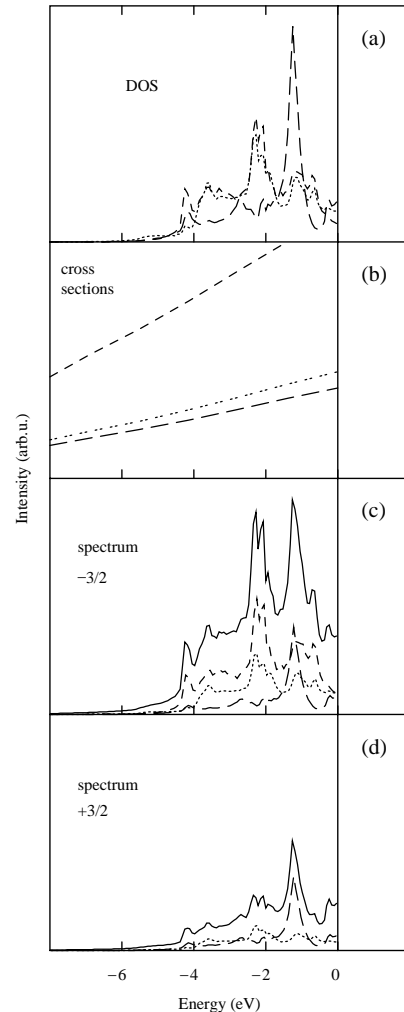


Fig. 3. Spin-down curves as in Fig. 2.

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References

- [1] G. Hörmandinger, P. Weinberger, P. Marksteiner, J. Redinger, Phys. Rev. B 38 (1988) 1040.
- [2] L. Szunyogh, P. Weinberger, J. Redinger, Phys. Rev. B 46 (1992) 2015.
- [3] P. Weinberger, L. Szunyogh, B.I. Bennett, Phys. Rev. B 47 (1993) 10154.
- [4] J. Minar, V. Popescu, H. Ebert, Phys. Rev. B 62 (2000) 10051.