Fully relativistic angle-resolved one-step theory of ultraviolet (inverse) photoemission for general nonlocal potentials

J. Braun[1], J. Minar[2], M. Donath[1] and H. Ebert[2]

Physikalisches Institut, Westfälische Wilhelms-Universität Münster, D-48149 Münster, Germany

Institut für Physikalische Chemie, Ludwig-Maximilians-Universität München, D-80333 München, Germany

MOTIVATION

An improved formulation of the one-step model of photoemission from crystal surfaces is proposed which overcomes different limitations of the original theory. Considering the results of an electronic-structure calculation, the electronic (one-particle) potential and the (many-body) self-energy, as given quantities, we derive explicit expressions for the dipole transition-matrix elements. The theory is formulated within a spin-polarized, relativistic framework for general nonspherical and space-filling one-particle potentials and general nonlocal, complex and energy-dependent self-energies. It applies to semi-infinite lattices with perfect lateral translational invariance and arbitrary number of atoms per unit cell.

Fermi's golden rule

$$\Gamma = -\frac{2\pi}{\hbar} \,|<\Psi_F|\Delta|\Psi_I>|^2\, \delta(E_F - E_I - e_{ph})$$

$$\Delta^{PES} = \sum_{\mathbf{e},\mathbf{k}} M^P_{\mathbf{e},\mathbf{k}}\, a^\dagger_{\mathbf{e}} a_{\mathbf{k}}$$

$$\Delta^{IPE} = \sum_{\mathbf{e},\mathbf{k}} M^P_{\mathbf{k},\mathbf{e}}\, a^\dagger_{\mathbf{k}} a_{\mathbf{e}}$$

$$M^P_{\mathbf{e},\mathbf{k}} = <\phi^{SP}_{\mathbf{e}}|\mathbf{A_o}\cdot\boldsymbol{\alpha}|\phi_{\mathbf{k}}>$$

Sudden approximation

The interaction of the photoelectron with the rest system is neglected

PES: $|\Psi_I> = |\Psi^N_0>$

PES: $|\Psi_F> = a^\dagger_{\mathbf{e}}|\Psi^{N-1}_S>$

IPE: $|\Psi_I> = a^\dagger_{\mathbf{e}}|\Psi^N_0>$

IPE: $|\Psi_F> = |\Psi^{N+1}_S>$

Inserting $|\Psi_I>$ and $|\Psi_F>$ in Fermi's golden rule
Summation over all possible final states
Averaging in the Grand Canonical Ensemble

$$\frac{1}{2\pi} < [T^\dagger(t), T(t')]_+ > = A^{(1)}(t,t') = \frac{1}{2\pi\hbar}\int dE e^{-\frac{i}{\hbar}E(t-t')}\, \mathbf{A}^{(1)}(\mathbf{E})$$

$$T^{PES} = \sum_{\mathbf{k}} M^P_{\mathbf{e},\mathbf{k}}\, a_{\mathbf{k}} \qquad T^{IPE} = \sum_{\mathbf{k}} M^P_{\mathbf{e},\mathbf{k}}\, a^\dagger_{\mathbf{k}}$$

One-step model of photoemission

$$I(e_e, \mathbf{k}_\|) = \int d\mathbf{r} \int d\mathbf{r}'\, \Psi^\dagger_e(\mathbf{r})\, \mathbf{A}_o\boldsymbol{\alpha}\, A^{(1)}(\mathbf{r},\mathbf{r}',E)\, \mathbf{A}_o\boldsymbol{\alpha}^\dagger\, \Psi_e(\mathbf{r}')$$

Dipol operator

$$<\Psi_f|\mathbf{A}_o\boldsymbol{\alpha}|\Psi_i> \sim \left(\mathbf{A}_0\boldsymbol{\nabla} + \frac{i\omega}{c}\boldsymbol{\alpha}\mathbf{A}_0\right) V_{\rm LDA}(\mathbf{r}) + (\mathbf{A}_0\boldsymbol{\nabla})\,\beta\boldsymbol{\sigma}\mathbf{B}_{\rm LDA}(\mathbf{r}) + \frac{\omega}{c}\beta\mathbf{A}_0\times\boldsymbol{\sigma}\mathbf{B}_{\rm LDA}(\mathbf{r})$$

Dipol selection rules

$$M \neq 0 \;\; for \;\; B_z : \delta_{\kappa,\kappa'\,(-\kappa'-1)}\delta_{\mu,\mu'} \quad B_{xy} : \delta_{\kappa,\kappa'\,(-\kappa'-1)}\delta_{\mu,\mu'\pm1}$$

INITIAL- AND FINAL STATES

Relativistic LDA-Hamiltonian

$$h_{\rm LDA}(\mathbf{r}) = -ic\boldsymbol{\alpha}\boldsymbol{\nabla} + \beta c^2 - c^2 + V_{\rm LDA}(\mathbf{r}) + \beta\boldsymbol{\sigma}\mathbf{B}_{\rm LDA}(\mathbf{r})$$

$$V_{\rm LDA}(\mathbf{r}) = \frac{1}{2}(V^\uparrow_{\rm LDA}(\mathbf{r}) + V^\downarrow_{\rm LDA}(\mathbf{r})) \qquad \mathbf{B}_{\rm LDA}(\mathbf{r}) = \frac{1}{2}(V^\uparrow_{\rm LDA}(\mathbf{r}) - V^\downarrow_{\rm LDA}(\mathbf{r}))\mathbf{b}$$

Generalized nonlocal potential

$$U(\mathbf{r},\mathbf{r}',E) = \delta(\mathbf{r}-\mathbf{r}')\,(V_{\rm LDA}(\mathbf{r}) + \beta\boldsymbol{\sigma}\mathbf{B}_{\rm LDA}(\mathbf{r})) + V(\mathbf{r},\mathbf{r}',E) + \beta\boldsymbol{\sigma}\mathbf{B}(\mathbf{r},\mathbf{r}',E)$$

$$V(\mathbf{r},\mathbf{r}',E) = \frac{1}{2}(\Sigma^\uparrow(\mathbf{r},\mathbf{r}',E) + \Sigma^\downarrow(\mathbf{r},\mathbf{r}',E)) \qquad \mathbf{B}(\mathbf{r},\mathbf{r}',E) = \frac{1}{2}(\Sigma^\uparrow(\mathbf{r},\mathbf{r}',E) - \Sigma^\downarrow(\mathbf{r},\mathbf{r}',E))\,\mathbf{b}$$

Dyson equation for the initial state Green function

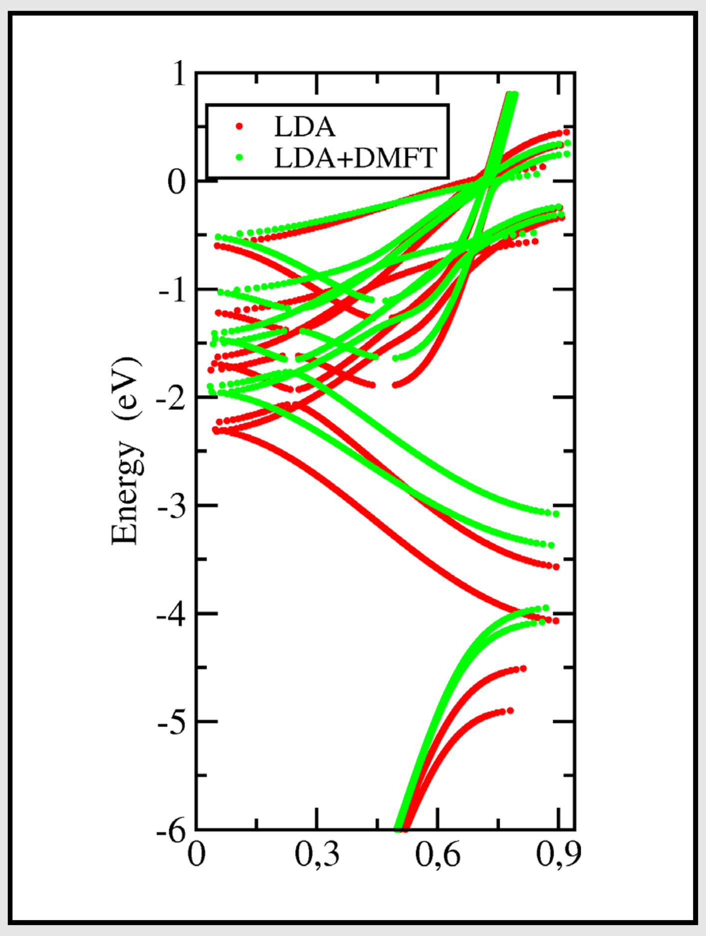
$$\left[E + \mu_0 + ic\boldsymbol{\alpha}\boldsymbol{\nabla} - \beta c^2 + c^2\right] G^+_1(\mathbf{r},\mathbf{r}',E) + \int U(\mathbf{r},\mathbf{r}'',E) G^+_1(\mathbf{r}'',\mathbf{r}',E)\, d\mathbf{r}'' = \delta(\mathbf{r}-\mathbf{r}')$$

Time reversed SPLEED state for the photoelectron

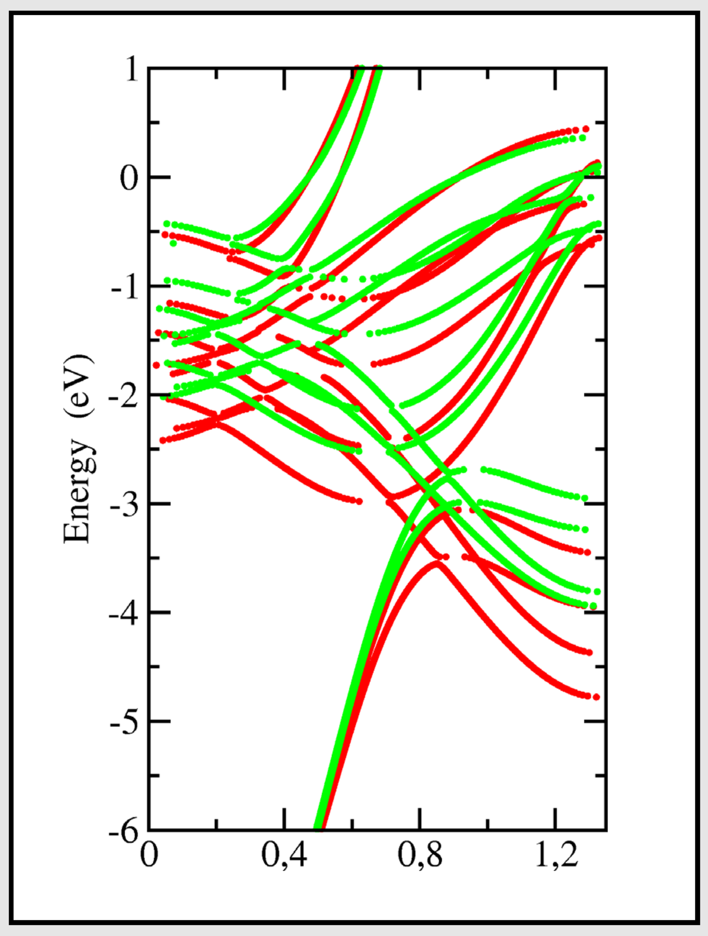
$$\phi^{SP}_e(\mathbf{r}) \equiv \langle \mathbf{r}|G^-_2|e,\mathbf{k}_\|\rangle$$

Bandstructures for different magnetization directions

Ni(100) Γ-Δ-X for $M_\perp$



Ni(110) Γ-Σ-X for $M_\|$

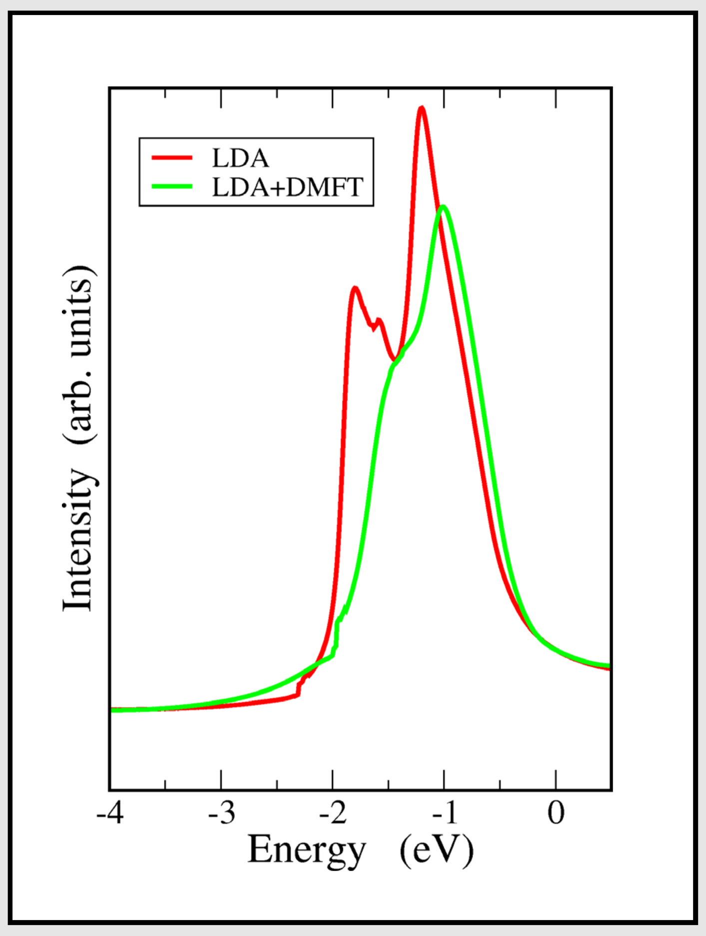


$$U_{\rm LDA}(\mathbf{r}) = V_{\rm LDA}(\mathbf{r}) + \mathbf{B}_{\rm LDA}(\mathbf{r})$$

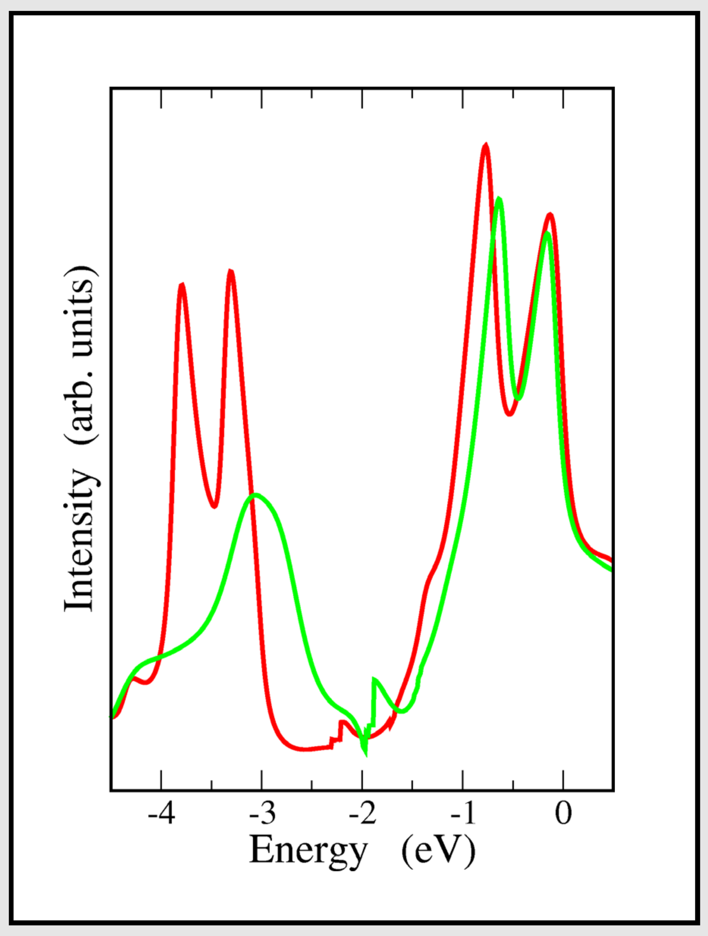
$$U^{\rm DMFT}_{\rm LDA}(\mathbf{r},E) = V_{\rm LDA}(\mathbf{r}) + \mathbf{B}_{\rm LDA}(\mathbf{r}) + V^{\rm DMFT}(E) + \mathbf{B}^{\rm DMFT}(E)$$

normal emission, $M_\perp$ and $M_\|$ to the surface, $\hbar\omega$=21.2 eV

Ni(100) Γ-Δ-X for $M_\perp$



Ni(110) Γ-Σ-X for $M_\|$



OUTLOOK

Numerical implementation of the complete one-step theory of nonlocal photoemission spectroscopy for ordered and disordered systems.

Generalization of the one-step model of nonlocal photoemission spectroscopy to low dimensional systems.

References

[1] J. B. Pendry, *Low Energy Electron Diffraction*, (Academic, London, 1974).
[2] J. B. Pendry, Surf. Sci. **57**, 679 (1976).
[3] J. F. L. Hopkinson, J. B. Pendry, and D. J. Titterington, Comput. Phys. Commun. **19**, 69 (1980).

[4] B. Ginatempo, P. J. Durham, and B. I. Gyorffy, J. Phys. C **1**, 6483 (1983); J. Braun, G. Thörner, and G. Borstel, phys. stat. sol. (b) **130**, 643 (1985); and **144**, 609 (1987); S. V. Halilov, E. Tamura, H. Gollisch, D. Meinert, and R. Feder, J. Phys.: Condens. Matter **5**, 3859 (1993); J. Henk, S. V. Halilov, T. Scheunemann, and R. Feder, Phys. Rev. B **50**, 8130 (1994); M. Fluchtmann, M. Graß, J. Braun, and G. Borstel, Phys. Rev. B **95**, 9564 (1995).
[5] J. Braun, Rep. Prog. Phys. **59**, 1267 (1996).
[6] C. Meyer, M. Potthoff, W. Nolting, G. Borstel and J. Braun, phys. stat. sol. (b) **203**, 441 (1997).