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Relativistic Green function theory of layer densities of states and photoemission from magnetic compounds

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Abstract. To study the electronic structure of ferromagnetic crystalline compounds, their surfaces and adsorbed ultrathin films, a fully relativistic Green function formalism has been developed. For a semi-infinite system, which is described by a complex effective potential, the single-particle Green function and thence the layer-, $k_{\parallel}$- and spin- or symmetry-resolved densities of states—at the surface and in the bulk—are obtained using a layer Korringa–Kohn–Rostoker method. This Green function—for quasi-hole states—is employed in a relativistic one-step model formalism to yield spin- and angle-resolved photoemission intensities.

1. Introduction

Over the past decade, a strongly increasing number of both experimental and theoretical investigations have firmly established spin-resolved photoemission spectroscopy as a powerful tool for studying in detail the electronic structure of ferromagnetic and non-magnetic crystals, their surfaces and ultrathin films adsorbed on them; see, for example the reviews by Feder (1985), Kirschner (1985), Kisker (1987) and Heinzmann (1990), and a small selection of original articles by Kezzler *et al* (1987), Tamura *et al* (1987, 1989), Schneider *et al* (1989, 1991), Ginatempo *et al* (1989), Ginatempo and Gyorffy (1990), Gollisch and Feder (1990), Hillbrecht *et al* (1990), Stoppmanns *et al* (1991), Tamura and Feder (1991a, b), Koenig *et al* (1991) and Braun *et al* (1991), and the references therein. Maximal information can be obtained if experimental data are analysed and interpreted with the aid of realistic numerical calculations. To this end, fully relativistic one-step model photoemission formalisms have been developed for magnetic and non-magnetic elemental solids (Ackermann and Feder 1985a, b, Braun *et al* 1985, 1987) and for non-magnetic elemental and compound solids (Ginatempo *et al* 1985, 1989). These theories have in common that they are essentially generalizations of Pendry's (1976) non-relativistic layer Korringa–Kohn–Rostoker (KKR) photoemission theory, but they differ in formal details and in computational implementations. In recent applications (Stoppmanns *et al* 1991, Tamura and Feder 1991a, b) it has been demonstrated that additional information can be gained if the spin-resolved photoemission calculations are accompanied by relativistic layer-resolved density of states (LDOS) calculations using a Green function formalism developed by Tamura and Feder (1989) as an extension of the non-relativistic method of Hora and Scheffler (1984).

In this paper, we present a coherent view of relativistic Green function theory of layer density of states and photoemission for non-magnetic and ferromagnetic systems with several

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generalizations beyond previous work. In particular, we focus on the following aspects. The layer density of states method for systems with one atom per unit cell, which was only briefly outlined by Tamura and Feder (1989) and Tamura (1992a, b), is described in more detail and extended to the case of several atoms per unit cell. The underlying Green function is extended to connect different atomic layers. It is then employed to provide an alternative evaluation of the photoemission density matrix. This new form as well as the form dating back to Ackermann and Feder (1985a, b) are extended to handle ferromagnetic compounds.

Our paper is organized as follows. Section 2 is devoted to the relativistic single-particle Green function for semi-infinite crystalline ferromagnetic compounds including ultrathin overlayer films. In section 3, the corresponding layer-, spin- and symmetry-resolved densities of states are evaluated. Section 4 deals with spin-resolved photoemission.

2. Layer Green function for the half-space crystalline system

As the basis for treating one-electron excitations in a ferromagnet, we use a Dirac Hamiltonian, in which the complex self-energy matrix is approximated by an effective electrostatic potential $V(E,\boldsymbol{r})$ and an effective magnetic field $\boldsymbol{B}(E,\boldsymbol{r})$ (cf Feder 1985 section 4.2.1. and references therein):

$$\hat{H} = c\hat{\alpha}\cdot\boldsymbol{p} + \hat{\beta}mc^2 + V(E,\boldsymbol{r}) - \hat{\beta}\hat{\sigma}\cdot\boldsymbol{B}(E,\boldsymbol{r}) \tag{1}$$

where c is the velocity of light, m the electron rest mass ($m = 1$ is supposed), with the standard representation

$$\hat{\alpha} = \begin{pmatrix} 0 & \hat{\sigma} \\ \hat{\sigma} & 0 \end{pmatrix} \qquad \hat{\beta} = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

Spin–orbit coupling and ferromagnetic exchange coupling are thus treated on an equal footing. Non-magnetic systems are described by equation (1) with $\boldsymbol{B}(E,\boldsymbol{r}) = 0$.

In the following, we specialize to a semi-infinite crystal model, which may be viewed as a stack of atomic layers extending infinitely in the two dimensions (x, y) parallel to the surface, with the topmost layer(s) possibly consisting of atoms of a different species. The potential and the magnetic field are assumed to be constant between touching muffin-tin spheres, but may have non-spherical contributions within the spheres. The transition from the constant inner potential to the vacuum (taken in negative z direction), the surface potential barrier, is assumed to have some smooth form with image-potential asymptotics.

The single-particle Green function (GF) (4×4) matrix G^{tot} could in principle be evaluated from a Dyson equation involving the complete interaction term of equation (1) and the vacuum Green function G^0 associated with the free part $\hat{H}_0$ of our Hamiltonian:

$$\sum_l [E - \hat{H}_0]_{il} G^0_{lk}(\boldsymbol{r},\boldsymbol{r}',E) = \delta(\boldsymbol{r}-\boldsymbol{r}')\delta_{ik} \qquad i,k = 1,2,3,4 \tag{2}$$

where i, k denote bispinor indices and $\hat{H}_0 = c\hat{\alpha}\cdot\boldsymbol{p} + \hat{\beta}c^2$.

As was proposed by Hora and Scheffler (1984) within a non-relativistic framework, it is however more practicable to split the potential into a contribution from the layer under consideration and the remainder. This leads to a Dyson equation involving a so-called empty-layer Green function G^{EL}. As the first step, we now choose the vacuum GF G^0 in a way corresponding to a hypothetical empty lattice (with the same geometry as the actual

one). Within an empty monolayer, this vacuum (empty-space) Green function G^{ES} thus has to satisfy the two-dimensional Bloch theorem

$$G^{ES}(\boldsymbol{r}+\boldsymbol{R},\boldsymbol{r}',E,\boldsymbol{k}_\parallel)=\exp(\mathrm{i}\boldsymbol{k}_\parallel\boldsymbol{R})G^{ES}(\boldsymbol{r},\boldsymbol{r}',E,\boldsymbol{k}_\parallel)$$

and can be constructed from the non-relativistic empty-space GF (Hora and Scheffler 1984, equations (6*a*, *b*))

$$G^{ES}_{\text{nonrel}}(\boldsymbol{r},\boldsymbol{r}',E,\boldsymbol{k}_\parallel)=-\frac{1}{4\pi}\sum_{\boldsymbol{R}}\frac{\exp(\mathrm{i}k|\boldsymbol{r}-\boldsymbol{r}'-\boldsymbol{R}|)}{|\boldsymbol{r}-\boldsymbol{r}'-\boldsymbol{R}|}\exp(\mathrm{i}\boldsymbol{k}_\parallel\boldsymbol{R}) \tag{3}$$

by the following operation:

$$G^{ES}(\boldsymbol{r},\boldsymbol{r}',E,\boldsymbol{k}_\parallel)=(1/c^2)[E+\hat{H}_0]G^{ES}_{\text{nonrel}}(\boldsymbol{r},\boldsymbol{r}',E,\boldsymbol{k}_\parallel). \tag{4}$$

Here $\boldsymbol{R}$ is the two-dimensional lattice vector and $k\equiv[(E^2-c^4)/c^2]^{1/2}$. It is supposed in equation (3) that $\boldsymbol{r}$ and $\boldsymbol{r}'$ are positioned in the same empty layer. In the case of $\boldsymbol{r}$ and $\boldsymbol{r}'$ positioned in different empty layers N and N' it will be useful to define the GF in the following form:

$$G^{ES}_{\text{nonrel}}(\boldsymbol{r}_{nN},\boldsymbol{r}'_{n'N'},E,\boldsymbol{k}_\parallel)=-\frac{1}{4\pi}\sum_{\boldsymbol{R}_{N,N'}}\frac{\exp(\mathrm{i}k|\boldsymbol{r}_{nN}-\boldsymbol{r}_{n'N'}-\boldsymbol{R}_{N,N'}|)}{|\boldsymbol{r}_{nN}-\boldsymbol{r}_{n'N'}-\boldsymbol{R}_{N,N'}|}\exp(\mathrm{i}\boldsymbol{k}_\parallel\boldsymbol{R}_{N,N'}) \tag{3a}$$

where $\boldsymbol{r}_{nN}\equiv\boldsymbol{r}+\boldsymbol{c}_{nN}+\boldsymbol{R}_N$, $\boldsymbol{r}'_{n'N'}\equiv\boldsymbol{r}'+\boldsymbol{c}'_{n'N'}+\boldsymbol{R}'_{N'}+\boldsymbol{d}_{N,N'}$, $\boldsymbol{d}_{N,N'}$ is an interlayer displacement vector, $\boldsymbol{c}_{nN}$ is the nth basis atom vector inside the unit cell of the Nth layer and $\boldsymbol{R}_{N,N'}\equiv\boldsymbol{R}_N-\boldsymbol{R}'_{N'}$ is the difference between two-dimensional lattice vectors of the Nth and N'th layers. The relativistic G^{ES} obtained by substituting equation (3*a*) into equation (4) satisfies the two-diemsnional Bloch condition in both empty layers.

In view of employing planar boundary conditions we transform equation (4) into the plane-wave representation (with the layer and atomic basis indices omitted):

$$G^{ES}(\boldsymbol{r},\boldsymbol{r}',E,\boldsymbol{k}_\parallel)=\sum_{ss'}\sum_{\substack{gg'\\ \tau\tau'}}\lambda^{ss'}_{\substack{gg'\\ \tau\tau'}}u^s_{g\tau}\exp(\mathrm{i}\boldsymbol{k}^s_g\boldsymbol{r})\otimes(u^s)^{\mathrm{T}}_{g'\tau'}\exp(-\mathrm{i}\boldsymbol{k}^s_{g'}\boldsymbol{r}')\delta_{s,\mathrm{sgn}(z-z')} \tag{5}$$

$$\lambda^{ss'}_{\substack{gg'\\ \tau\tau'}}\equiv\frac{1}{2\mathrm{i}S_{\text{cell}}(k^z_g)^+}\frac{E+c^2}{c^2}\delta_{ss'}\delta_{gg'}\delta_{\tau\tau'} \tag{6}$$

$$u^s_{g\tau}\equiv\begin{pmatrix}\chi_\tau\\ [c\boldsymbol{\sigma}\cdot\boldsymbol{k}^s_g/(E+c^2)]\chi_\tau\end{pmatrix}\qquad(u^s)^{\mathrm{T}}_{g'\tau'}\equiv\begin{pmatrix}\chi^{\mathrm{T}}_{\tau'} & \dfrac{c\boldsymbol{\sigma}\cdot\boldsymbol{k}^s_{g'}}{E+c^2}\chi^{\mathrm{T}}_{\tau'}\end{pmatrix} \tag{7}$$

where $u^s_{g\tau}$ and $(u^s)^{\mathrm{T}}_{g\tau}$ can be considered as amplitudes of the waves $\exp(\mathrm{i}\boldsymbol{k}^s_g\boldsymbol{r})$ and $\exp(-\mathrm{i}\boldsymbol{k}^s_g\boldsymbol{r}')$, respectively, $s\equiv\pm$, χ_τ are the two-component Pauli spinors, $\tau=1,2$, $\boldsymbol{k}^s_g\equiv\{\boldsymbol{k}_\parallel+\boldsymbol{g},\ \mathrm{sgn}(s)k^z_g\}\equiv\{\boldsymbol{k}^\parallel_g,\ (k^z_g)^s\}$ and $\boldsymbol{g}$ is the vector of the two-dimensional lattice reciprocal to the one defined by the $\boldsymbol{R}_{N,N'}$.

So far we have focused on the empty-space GF. Now we consider a semi-infinite system with two monolayers removed such that in general there is some finite number (slab) of monolayers in between (see figure 1(*a*)). For the general case of several atoms per two-dimensional unit cell we mean by 'monolayer' a layer infinite in the xy plane (parallel to the surface plane) and of a finite z thickness corresponding to the unit cell. One of the empty

monolayers (cf figure 1(*a*)) contains z' (the z component of the Green function argument r'), the other z. For this sytem, the empty-space GF is modified by a boundary correction G^{B} to yield the 'two-empty-layer Green function' G^{EL}:

$$G^{EL} = G^{ES} + G^{B}. \tag{8}$$

To evaluate G^{B}, we fix the z' plane and consider local boundary conditions at the z plane, which are via simple free-propagation phase factors equivalent to the boundary conditions at the planes bounding the empty monolayer. These local boundary conditions imply back-scattering of plane waves and can be described by local reflection matrices R^{+-}_{loc} and R^{-+}_{loc} at planes $z-\epsilon$ and $z+\epsilon$ ($\epsilon \to +0$), respectively (cf figure 1(*b*)). There are two possible situations for the scattering of the initial wave $u^{+}_{g\tau}\exp[i(k^{z}_{g})^{+}z]$ (cf equation (5)), which is going in the $+z$ direction at $z > z'$ (see figure 1(*b*)), which is an enlarged part of figure 1(*a*)): (i) the scattered wave $u^{-}_{g\tau}\exp[i(k^{z}_{g})^{-}z]$ moves in the $-z$ direction (solid bold line in figure 1(*b*)), and (ii) the scattered wave $u^{+}_{g\tau}\exp[i(k^{z}_{g})^{+}z]$ moves in the $+z$ direction (dashed bold line in figure 1(*b*)).

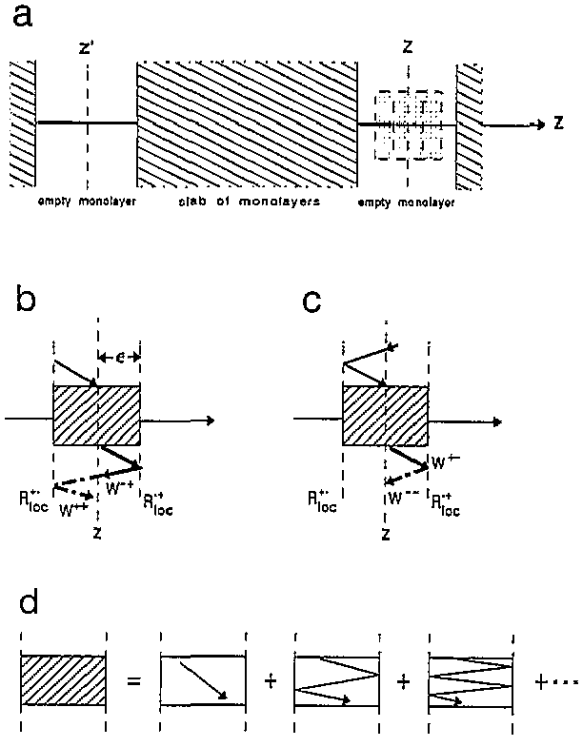

Figure 1. Illustration of the boundary correction part G^{B} of the empty-layer Green function G^{EL} (cf equations (8) and (9)). (*a*) Sketch of a semi-infinite system with two monolayers (parallel to the surface) removed. (*b*) Enlarged view of the dotted area in the right-hand empty layer of part (*a*). The upper right-pointing solid arrow stands for the original wavefield. The hatched area symbolizes the multiple scattering series due to the local reflection matrices R^{+-}_{loc} and R^{-+}_{loc}. The left-pointing solid arrow and the right-pointing dashed arrow correspond to scattered waves with amplitudes W^{-+} and W^{++} as given by equations (9) and (9*a*), respectively. (*c*) Analogous to part (*b*) but for the source monolayer (containing z') to the right of the empty monolayer containing z. The scattering amplitudes W^{+-} and W^{--} are given by equations (9*b*) and (9*c*), respectively. (*d*) Multiple scattering series as used in parts (*b*) and (*c*).

The hatched area in figure 1(*b*) represents the geometrical series of the multiple scattering between R^{+-}_{loc} and R^{-+}_{loc}, which can be evaluated as $[1 - R^{+-}_{loc}R^{-+}_{loc}]^{-1}$. Eventually we get the following expression for the empty-layer GF (equation (8)) for the situation of figure 1(*a*) ($z > z'$):

$$G^{EL}(r, r', E, k_{\parallel}) = \sum_{\substack{gg'\\ \tau\tau'}} \Big[\lambda^{++}_{\substack{gg'\\ \tau\tau'}} u^{+}_{g\tau}\exp(ik^{+}_{g}r) \otimes (u^{+})^{T}_{g'\tau'}\exp(-ik^{+}_{g'}r')$$

$$+ W^{-+}_{\substack{gg'\\ \tau\tau'}} u^{-}_{g\tau} \exp(\mathrm{i}k^{-}_{g} r) \otimes (u^{+})^{\mathrm{T}}_{g'\tau'} \exp(-\mathrm{i}k^{+}_{g'} r')$$

$$+ W^{++}_{\substack{gg'\\ \tau\tau'}} u^{+}_{g\tau} \exp(\mathrm{i}k^{+}_{g} r) \otimes (u^{+})^{\mathrm{T}}_{g'\tau'} \exp(-\mathrm{i}k^{+}_{g'} r')\Big] \tag{8a}$$

where the amplitudes of the scattered waves are

$$W^{-+}_{\substack{gg'\\ \tau\tau'}} \equiv (R^{-+}_{\mathrm{loc}}[1 - R^{+-}_{\mathrm{loc}} R^{-+}_{\mathrm{loc}}]^{-1})_{\substack{gg''\\ \tau\tau''}} \lambda^{++}_{\substack{g''g'\\ \tau''\tau'}} \tag{9}$$

$$W^{++}_{\substack{gg'\\ \tau\tau'}} \equiv (R^{+-}_{\mathrm{loc}} W^{-+})_{\substack{gg'\\ \tau\tau'}}. \tag{9a}$$

To determine the local reflection matrices, we require a scattering matrix for the 'slab' and matrices R^{+-} and R^{-+} describing reflection from the left-hand boundary plane of the first empty layer and the right-hand boundary plane of the second layer, respectively (cf figure 1(*a*)). If the surface (not shown) is towards the left, R^{-+} is − except for z in the first few layers—the bulk reflection matrix. For the special case of the slab consisting only of one (filled) monolayer, we find (in matrix form):

$$R^{+-}_{\mathrm{loc}} = \tilde{M}^{+-} + \tilde{M}^{++}[1 - \tilde{R}^{+-}\tilde{M}^{-+}]^{-1}\tilde{R}^{+-}\tilde{M}^{--} \tag{10}$$

$$R^{-+}_{\mathrm{loc}} = \tilde{R}^{-+} \tag{10a}$$

where the tilde denotes the inclusion of the appropriate phase factors. The transmission/reflection matrices $M^{ss'}$ of the monolayer with several atoms per unit cell are:

$$M^{ss'}_{\substack{gg'\\ \tau\tau'}} = \frac{8\pi^2}{S_{\mathrm{cell}}|k^{s}_{g}|(k^{z}_{g})^{+}} \sum_{\substack{\kappa\mu n\\ \kappa'\mu' n'}} \mathrm{i}^{-l'} t^{n}_{\substack{\kappa'\mu'\\ \kappa\mu}} C(l'\tfrac{1}{2}j';\mu'-\tau,\tau) Y^{\mu'-\tau}_{l'}(k^{s}_{g})$$

$$\times \sum_{\kappa''\mu''} \mathrm{i}^{l''} C(l''\tfrac{1}{2}j'';\mu''-\tau',\tau')(-1)^{-\mu''+\tau'} Y^{-\mu''+\tau'}_{l''}(k^{s'}_{g'})[1 - \gamma \hat{A}^{\mathrm{rel}}\hat{t}]^{-1}_{\substack{\kappa\mu n\\ \kappa''\mu'' n'}}$$

$$\times \exp(\mathrm{i}k^{s}_{g} r_{n} - \mathrm{i}k^{s'}_{g'} r_{n'})$$

with $\gamma \equiv -c^2/[\mathrm{i}k(E + c^2)]$. Here $\kappa\mu$ denote the usual relativistic angular momentum quantum numbers, r_n is the position of the nth atom within the layer unit cell, $t^{n}_{\substack{\kappa'\mu'\\ \kappa\mu}}$ is a usual single-site scattering t-matrix, and $A^{\mathrm{rel}}_{\substack{\kappa\mu n\\ \kappa'\mu' n'}}$ denotes the relativistic two-dimensional KKR structure constants. For a slab of several monolayers, the diffraction matrices $M^{ss'}$ in equation (10) refer to the slab and are obtained by combining the diffraction matrices of the individual constituent layers.

Before obtaining the total GF for the half-space crystalline system it is convenient to transform the empty-layer G^{EL} from the plane-wave representation (8*a*) to the spherical-wave representation

$$G^{\mathrm{EL}}(r_{nN}, r'_{n'N'}, E, k_{\parallel}) = G^{\mathrm{ES}} + G^{\mathrm{B}} = G_0 \delta_{N,N'}\delta_{n,n'} + (G^{\mathrm{A}} + G^{\mathrm{B}})$$

$$= G_0 \delta_{N,N'}\delta_{n,n'} + \sum_{\substack{\kappa\mu\\ \kappa'\mu'}} D^{NN'}_{\substack{\kappa\mu n\\ \kappa'\mu' n'}} \begin{pmatrix} j_l(kr)|\chi_{\kappa\mu}\rangle \\ \mathrm{i}\,\mathrm{sgn}(\kappa) v j_{\bar{l}}(kr)|\chi_{\bar{\kappa}\mu}\rangle \end{pmatrix}_{nN}$$

$$\otimes \left[j_{l'}(kr')\langle\chi_{\kappa'\mu'}|, -\mathrm{i}\,\mathrm{sgn}(\kappa) v j_{\bar{l}'}(kr')\langle\chi_{\bar{\kappa}'\mu'}| \right]_{n'N'} \tag{11}$$

where $\bar{l} \equiv l - (\kappa/|\kappa|)$, $\bar{\kappa} \equiv -\kappa$, $\nu \equiv ck/(E + c^2)$ and $|\chi_{\kappa\mu}\rangle$ means the Pauli central-field spinor. G_0 is the single-site (irregular) part of the empty-space GF:

$$G_0(\boldsymbol{r}_{nN}, \boldsymbol{r}'_{nN}) = -ik \begin{cases} \sum_{\kappa\mu} \langle r_{nN} \mid h^{(1)}_{\kappa\mu} \rangle \langle j_{\kappa\mu} \mid r'_{nN} \rangle & \text{for } r > r' \\ \sum_{\kappa\mu} \langle r_{nN} \mid j_{\kappa\mu} \rangle \langle h^{(1)}_{\kappa\mu} \mid r'_{nN} \rangle & \text{for } r < r' \end{cases}$$

and G^{A} (in equation (11)) is its regular part. G^{A} and G^{B} are combined to produce $D^{NN'}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}} = A^{NN'\mathrm{rel}}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}} + B^{NN'}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}}$, where the structure constants $A^{NN'\mathrm{rel}}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}}$ (referred to the lattice defined by the vectors $\boldsymbol{d}_{NN'} + \boldsymbol{R}_{NN'}$) are

$$A^{NN'\mathrm{rel}}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}} = \frac{1}{\gamma} \sum_{\boldsymbol{R}_{N,N'}} G^{\mathrm{rel}}_{\kappa'\mu',\kappa\mu}(\boldsymbol{c}_{nN} - \boldsymbol{c}_{n'N'} - \boldsymbol{d}_{N,N'} - \boldsymbol{R}_{N,N'}) \exp(i\boldsymbol{k}_{\parallel}\boldsymbol{R}_{N,N'})(1 - \delta_{0,\boldsymbol{R}_N}\delta_{nN,n'N'}) \tag{12}$$

where $G^{\mathrm{rel}}_{\kappa'\mu',\kappa\mu}$ is defined as

$$G^{\mathrm{rel}}_{\kappa'\mu',\kappa\mu}(\boldsymbol{c}_{nN} - \boldsymbol{c}_{n'N'} - \boldsymbol{d}_{N,N'} - \boldsymbol{R}_{N,N'}) = \sum_{\tau} G^{\mathrm{nonrel}}_{l'\mu'-\tau',l\mu-\tau}(\boldsymbol{c}_{nN} - \boldsymbol{c}_{n'N'} - \boldsymbol{d}_{N,N'} - \boldsymbol{R}_{N,N'})$$
$$\times C(l\tfrac{1}{2}j; \mu - \tau, \tau) C(l'\tfrac{1}{2}j'; \mu' - \tau', \tau')$$

and non-relativistic structure constants are

$$G^{\mathrm{nonrel}}_{l'\mu'-\tau',l\mu-\tau}(\boldsymbol{c}_{nN} - \boldsymbol{c}_{n'N'} - \boldsymbol{d}_{N,N'} - \boldsymbol{R}_{N,N'})$$
$$= \sum_{L''} 4\pi i^{l-l'-l''}(-1)^{m''+m'} h^{(1)}_{l''}(k|\boldsymbol{c}_{nN} - \boldsymbol{c}_{n'N'} - \boldsymbol{d}_{N,N'} - \boldsymbol{R}_{N,N'}|)$$
$$\times Y_{l''-m''}(\boldsymbol{c}_{nN} - \boldsymbol{c}_{n'N'} - \boldsymbol{d}_{N,N'} - \boldsymbol{R}_{N,N'}) \int Y_L Y_{L''} Y_{l-m'}\, \mathrm{d}\Omega.$$

For the amplitudes of the spherical waves $B^{NN'}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}}$ scattered by the planar boundaries, we obtain (cf equation (8*a*) in the case $z > z'$):

$$B^{NN'}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}} = \sum_{\substack{ss' \\ g\tau, g'\tau'}} W^{ss'}_{\substack{gg' \\ \tau\tau'}} 16\pi^2 i^{l-l'} C(l\tfrac{1}{2}j; \mu - \tau, \tau) C(l'\tfrac{1}{2}j'; \mu' - \tau', \tau') Y_l^{-\mu+\tau}(k_g^s) Y_{l'}^{\mu'-\tau'}(k_{g'}^{s'}) \tag{13}$$

where for the case $z > z'$ we have non-zero only W^{-+} and W^{++} (equations (9) and (9*a*)) and for the case $z < z'$ only W^{+-} and W^{--} (cf figure 1(*c*)), which are

$$W^{+-}_{\substack{gg' \\ \tau\tau'}} \equiv ([1 - R^{+-}_{\mathrm{loc}} R^{-+}_{\mathrm{loc}}]^{-1} R^{+-}_{\mathrm{loc}})_{\substack{gg'' \\ \tau\tau''}} \lambda^{++}_{\substack{g''g' \\ \tau''\tau'}} \tag{9b}$$

$$W^{--}_{\substack{gg' \\ \tau\tau'}} \equiv (R^{-+}_{\mathrm{loc}} W^{+-})_{\substack{gg' \\ \tau\tau'}}. \tag{9c}$$

Now we can complete the derivation of the total GF G^{tot} for the half-space crystalline system—without empty monolayers—by means of the Dyson equation (with $\boldsymbol{k}_{\parallel}$ and E omitted):

$$G^{\mathrm{tot}}(\boldsymbol{r}_{nN},\boldsymbol{r}'_{n'N'}) = G^{\mathrm{EL}}(\boldsymbol{r}_{nN},\boldsymbol{r}_{n'N'}) + \int \mathrm{d}\tilde{\boldsymbol{r}}_N\, G^{\mathrm{EL}}(\boldsymbol{r}_{nN},\tilde{\boldsymbol{r}}_N)v(\tilde{\boldsymbol{r}}_N)G^{\mathrm{tot}}(\tilde{\boldsymbol{r}}_N,\boldsymbol{r}'_{n'N'}) + \int \mathrm{d}\tilde{\boldsymbol{r}}_{N'}\, G^{\mathrm{EL}}(\boldsymbol{r}_{nN},\tilde{\boldsymbol{r}}_{N'})v(\tilde{\boldsymbol{r}}_{N'})G^{\mathrm{tot}}(\tilde{\boldsymbol{r}}_{N'},\boldsymbol{r}'_{n'N'}) \tag{14}$$

where $v(\boldsymbol{r}_N)$ is the potential (a 4×4 matrix) in the Nth monolayer defined as $v(\boldsymbol{r}_N) = V(E,\boldsymbol{r}_N) - \hat{\beta}\hat{\sigma}\cdot\boldsymbol{B}(E,\boldsymbol{r}_N)$. The first (second) integration goes over the Nth (N'th) monolayer. It should be noted that equation (14) holds only if the monolayers N and N' are assumed to be different (and possibly non-equivalent).

The case of coinciding monolayers ($N = N'$) must be treated separately. The same-layer G^{tot} is needed first for evaluating the second integral in equation (14) and secondly to obtain the layer density of states. First the empty-space layer GF $G^{\mathrm{EL}}(\boldsymbol{r}_{nN},\boldsymbol{r}'_{n'N})$ is calculated as a combination of the two cases $(z-z')>0$ and $(z-z')<0$ in the limit $|z-z'|\to 0$, which obviously correspond to the half-crystalline system with only one (the Nth) monolayer removed. In this case one can formally use the expression (11) for $G^{\mathrm{EL}}(\boldsymbol{r}_{nN},\boldsymbol{r}'_{n'N})$ with the appropriate definition of $D^{NN}_{\substack{\kappa\mu n\\ \kappa'\mu'n'}}$ due to the change of $W^{ss'}_{\substack{gg'\\ \tau\tau'}}$. The same-layer total GF can then be obtained from the Dyson equation:

$$G^{\mathrm{tot}}(\boldsymbol{r}_{nN},\boldsymbol{r}'_{n'N}) = G^{\mathrm{EL}}(\boldsymbol{r}_{nN},\boldsymbol{r}_{n'N}) + \int \mathrm{d}\tilde{\boldsymbol{r}}_N\, G^{\mathrm{EL}}(\boldsymbol{r}_{nN},\tilde{\boldsymbol{r}}_N)v(\tilde{\boldsymbol{r}}_N)G^{\mathrm{tot}}(\tilde{\boldsymbol{r}}_N,\boldsymbol{r}'_{n'N}) \tag{14a}$$

and the suitable expansion of $G^{\mathrm{tot}}(\boldsymbol{r}_{nN},\boldsymbol{r}'_{n'N})$ in analogy with $G^{\mathrm{EL}}(\boldsymbol{r}_{nN},\boldsymbol{r}'_{n'N})$ (cf Tamura 1992b):

$$G^{\mathrm{tot}}(\boldsymbol{r}_{nN},\boldsymbol{r}'_{n'N}) = G_{\mathrm{a}}(\boldsymbol{r}_{nN},\boldsymbol{r}'_{n'N})\delta_{n,n'} + \sum_{\substack{\kappa\mu\\ \kappa'\mu'}} U^{NN}_{\substack{\kappa\mu n\\ \kappa'\mu'n'}} \begin{pmatrix} \psi^{\mathrm{R}}_{\kappa\mu}(kr)|\chi_{\kappa\mu}\rangle \\ \mathrm{i}\phi^{\mathrm{R}}_{\kappa\mu}(kr)|\chi_{\bar{\kappa}\mu}\rangle \end{pmatrix}_{nN} \otimes \left[\psi^{\mathrm{R}}_{\kappa'\mu'}(kr')\langle\chi_{\kappa'\mu'}|, -\mathrm{i}\phi^{\mathrm{R}}_{\kappa'\mu'}(kr')\langle\chi_{\bar{\kappa}'\mu'}|\right]_{n'N} \tag{15}$$

where $G_{\mathrm{a}}(\boldsymbol{r}_{nN},\boldsymbol{r}'_{n'N})$ is the single-atom GF (cf Tamura 1992a) defined as

$$G_{\mathrm{a}}(\boldsymbol{r}_{nN},\boldsymbol{r}'_{nN}) = -\mathrm{i}k \begin{cases} \displaystyle\sum_{\kappa\mu}\langle \boldsymbol{r}_{nN} \mid \psi^{\mathrm{I}}_{\kappa\mu}\rangle\langle\psi^{\mathrm{R}}_{\kappa\mu} \mid \boldsymbol{r}'_{nN}\rangle & \text{for } r > r' \\ \displaystyle\sum_{\kappa\mu}\langle \boldsymbol{r}_{nN} \mid \psi^{\mathrm{R}}_{\kappa\mu}\rangle\langle\psi^{\mathrm{I}}_{\kappa\mu} \mid \boldsymbol{r}'_{nN}\rangle & \text{for } r < r' \end{cases} \tag{16}$$

with the boundary conditions for the Dirac bispinors $\langle \boldsymbol{r}_{nN} \mid \psi^{\mathrm{R}}_{\kappa\mu}\rangle$ from equation (24) and for $\langle \boldsymbol{r}_{nN} \mid \psi^{\mathrm{I}}_{\kappa\mu}\rangle$:

$$\langle \boldsymbol{r}_{nN} \mid \psi^{\mathrm{I}}_{\kappa\mu}\rangle \overset{r\geq R^{\mathrm{mt}}_{nN}}{\longrightarrow} \begin{pmatrix} h^{(1)}_l(kr)|\chi_{\kappa\mu}\rangle \\ \mathrm{sgn}(\kappa)(1/\gamma)h^{(1)}_{\bar{l}}(kr)|\chi_{\bar{\kappa}\mu}\rangle \end{pmatrix}. \tag{17}$$

The expansion coefficients $U^{NN}_{\substack{\kappa\mu n\\ \kappa'\mu'n'}}$, which are due to intra- and interlayer multiple scattering (boundary conditions), are

$$U^{NN}_{\substack{\kappa\mu n\\ \kappa'\mu'n'}} = \sum_{\substack{\kappa''\mu''n''}} [1-\gamma\hat{D}^{NN}\hat{t}]^{-1}_{\substack{\kappa\mu n\\ \kappa''\mu''n''}} D^{NN}_{\substack{\kappa''\mu''n''\\ \kappa'\mu'n'}} \tag{18}$$

where atomic summation n'' is done over the layer unit cell. The coefficients D^{NN} are given by the sum of the expressions of equations (12) and (13), specialized to $N = N'$. We now have the total GF for the case of $\boldsymbol{r}$ and $\boldsymbol{r}'$ positioned in the same (Nth) monolayer.

For $\boldsymbol{r}$ and $\boldsymbol{r}'$ in different layers N and N', we expand G^{tot} analogously to equation (15) but with $U^{NN}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}}$ replaced by $U^{NN'}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}}$, where $N \neq N'$. Substitution into the Dyson equation (14) together with G^{EL} from equation (11) leads to the following equation for $U^{NN'}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}}$:

$$U^{NN'}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}} = D^{NN'}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}} + \gamma \sum_{\kappa''\mu''n''} D^{NN}_{\substack{\kappa\mu n \\ \kappa''\mu'' n''}} \sum_{\bar{\kappa}\bar{\mu}} t^{N}_{\substack{\kappa''\mu''n'' \\ \bar{\kappa}\bar{\mu}n''}} U^{NN'}_{\substack{\bar{\kappa}\bar{\mu}n'' \\ \kappa'\mu' n'}} + \gamma \sum_{\kappa''\mu''n''} D^{NN'}_{\substack{\kappa\mu n \\ \kappa''\mu'' n''}} \sum_{\bar{\kappa}\bar{\mu}} t^{N'}_{\substack{\kappa''\mu''n'' \\ \bar{\kappa}\bar{\mu}n''}} U^{N'N'}_{\substack{\bar{\kappa}\bar{\mu}n'' \\ \kappa'\mu' n'}} \tag{19}$$

or eventually

$$U^{NN'}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}} = \sum_{\kappa''\mu''n''} [\hat{\mathrm{I}} - \gamma \hat{D}^{NN} \hat{t}^{N}]^{-1}_{\substack{\kappa\mu n \\ \kappa''\mu'' n''}} D^{NN'}_{\substack{\kappa''\mu''n'' \\ \bar{\kappa}''\bar{\mu}''\bar{n}''}} [\hat{\mathrm{I}} + \gamma \hat{t}^{N'} \hat{U}^{N'N'}]_{\substack{\bar{\kappa}''\bar{\mu}''\bar{n}'' \\ \kappa'\mu' n'}} \tag{20}$$

where the atomic summations n'' and $\bar{n}''$ are done over the Nth and N'th layer unit cells, respectively. We note that equation (20) actually gives $U^{NN'}$ explicitly, since the 'same-layer' $U^{N'N'}$ occurring on the right-hand side has already been determined by equation (18).

3. The $K_{\parallel}$-resolved layer density of states

It is rather straightforward now to evaluate the $\boldsymbol{k}_{\parallel}$-resolved layer (indeed local) density of states (LDOS) from equation (15) for the GF at coinciding monolayers ($N = N'$):

$$N_{nN}(\boldsymbol{k}_{\parallel}, E) = -\frac{1}{\pi} \operatorname{Tr} \operatorname{Im} G^{\mathrm{tot}}_{\tau\tau}(\boldsymbol{r}_{nN}, \boldsymbol{r}'_{n'N}, \boldsymbol{k}_{\parallel}, E) = -\frac{1}{\pi} \sum_{\kappa\mu\tau} \operatorname{Im} \langle \kappa\mu, \tau | G^{\mathrm{tot}} | \kappa\mu, \tau \rangle \tag{21}$$

where $|\kappa\mu, \tau\rangle$ is the spin-angular part of the eigenfunction of the free Dirac Hamiltonian:

$$|\kappa\mu, 1\rangle \equiv \begin{pmatrix} |\chi_{\kappa\mu}\rangle \\ 0 \end{pmatrix} \qquad |\kappa\mu, 2\rangle \equiv \begin{pmatrix} 0 \\ |\chi_{\bar{\kappa}\mu}\rangle \end{pmatrix}. \tag{22}$$

Substitution of equation (15) into (21) gives the following expression for the layer- and angle-resolved density of states:

$$N_{nN}(\boldsymbol{k}_{\parallel}, E) = \frac{1}{\pi} \frac{1}{\gamma^2} \sum_{\kappa\mu} \left[\operatorname{Re}(A^{N}_{\kappa\mu n} + B^{N}_{\kappa\mu n}) + \operatorname{Im} A^{N}_{\kappa\mu n} U^{NN}_{\substack{\kappa\mu n \\ \kappa\mu n}} \right] \tag{23}$$

where $U^{NN}_{\substack{\kappa\mu n \\ \kappa\mu n}}$ is defined by equation (18) and

$$A^{N}_{\kappa\mu n} \equiv \int_0^{R^{\mathrm{mt}}_n} \mathrm{d}r\, r^2 \left[\psi^{\mathrm{R}}_{\kappa\mu n}(r) \psi^{\mathrm{R}}_{\kappa\mu n}(r) + \phi^{\mathrm{R}}_{\kappa\mu n}(r) \phi^{\mathrm{R}}_{\kappa\mu n}(r) \right]$$

$$B^{N}_{\kappa\mu n} \equiv \int_0^{R^{\mathrm{mt}}_n} \mathrm{d}r\, r^2 \left[\psi^{\mathrm{R}}_{\kappa\mu n}(r) \psi^{\mathrm{I}}_{\kappa\mu n}(r) + \phi^{\mathrm{R}}_{\kappa\mu n}(r) \phi^{\mathrm{I}}_{\kappa\mu n}(r) \right]$$

with the following matching conditions for the radial parts of the regular and irregular (superscripts R and I) wavefunctions

$$\begin{pmatrix} \psi^{\mathrm{R}}_{\kappa\mu n} \\ \mathrm{i}\phi^{\mathrm{R}}_{\kappa\mu n} \end{pmatrix} \rightarrow \begin{pmatrix} j_l(kr) + \sum_{\kappa'\mu'} h^{(1)}_{l'}(kr) t^n_{\kappa'\mu'\kappa\mu} \\ \mathrm{sgn}(\kappa)(1/\gamma)\left[j_{\bar{l}}(kr) + \sum_{\kappa'\mu'} h^{(1)}_{\bar{l}'}(kr) t^n_{\kappa'\mu'\kappa\mu} \right] \end{pmatrix} \tag{24}$$

$$\begin{pmatrix} \psi^{\mathrm{I}}_{\kappa\mu n} \\ \mathrm{i}\phi^{\mathrm{I}}_{\kappa\mu n} \end{pmatrix} \rightarrow -\begin{pmatrix} j_l(kr) - \sum_{\kappa'\mu'} h^{(1)}_{l'}(kr)(t^n_{\kappa'\mu'\kappa\mu} - \delta_{\kappa'\mu'\kappa\mu}) \\ \mathrm{sgn}(\kappa)(1/\gamma)\left[j_{\bar{l}}(kr) - \sum_{\kappa'\mu'} h^{(1)}_{\bar{l}'}(kr)(t^n_{\kappa'\mu'\kappa\mu} - \delta_{\kappa'\mu'\kappa\mu}) \right] \end{pmatrix} \tag{24a}$$

at $r \geqslant R^{\mathrm{mt}}_n$, where R^{mt}_n is the muffin-tin radius of the nth atom.

If we transform the spin-angular $\kappa\mu$ representation of equation (21) into an (lms) representation, the LDOS N in equation (21) decomposes into N^+ and N^-, i.e. spin-resolved densities of states. In a ferromagnet these strictly correspond to majority and minority spin states, if spin–orbit coupling is neglected. For non-magnetic systems with spin–orbit coupling, we can—by using appropriate double group basis functions—decompose N into contributions from different double group symmetry types.

We point out once more that the layer dependence of the LDOS is determined by the boundary-reflection conditions through $U^{NN}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}}$ containing left- and right-hand side reflection matrices R^{+-} and R^{-+} (equation (18)). For monolayers of a certain type there is a typical common right-hand side matrix R^{-+}, while the left-hand side matrices R^{+-} depend also on the position of a given monolayer and essentially determine the development of the LDOS on going from the surface into the depth of the system. These reflection matrices are conveniently calculated by means of the normal-mode (Bloch-wave) method (yielding the bulk reflection matrix) and a layer T-matrix multiplication scheme.

4. Spin-resolved photoemission

We can now proceed to the calculation of the spin- and angle-resolved photocurrent. Our approach is in essence a relativistic generalization of Pendry's (1976) Schrödinger-equation-based one-step model formalism. We thus have to evaluate the lowest-order contributing diagram (the 'naked' triangle) obtained from non-equilibrium perturbation theory (cf Keldysh 1965, Caroli *et al* 1973). The individual electron and hole Green functions are renormalized, but vertex corrections are ignored. The hole enters via its spectral density function. The photocurrent is then described by a spin-density matrix ρ, which in the detection plane (vacuum) is completely represented by the following 2×2 matrix (in the Pauli spinor basis) (cf Ackermann 1985, Ackermann and Feder 1985a, b equation (25))

$$\rho_{\tau\tau'} = (1/2\mathrm{i})[\tilde{\rho}_{\tau\tau'} - \tilde{\rho}^*_{\tau'\tau}] \tag{25a}$$

where

$$\begin{aligned} \tilde{\rho}_{\tau\tau'}(k_\parallel, E) = -\frac{1}{\pi}k \int \mathrm{d}r \int \mathrm{d}r' \langle \psi^{\tau}_{k_\parallel E} \mid r\rangle \Delta_\omega(r) G^{\mathrm{tot}}(r, r', E - \hbar\omega, k_\parallel) \Delta^+_\omega(r') \langle r' \mid \psi^{\tau'}_{k_\parallel E}\rangle \\ + -\frac{1}{\pi}k \sum_{N,n} J_{\mathrm{cell},N} \sum_{N',n'} J_{\mathrm{cell},N'} \int \mathrm{d}r_{nN} \int \mathrm{d}r'_{n'N'} \langle \psi^{\tau}_{k_\parallel E} \mid r_{nN}\rangle \Delta_\omega(r_{nN}) \\ \times G^{\mathrm{tot}}(r_{nN}, r'_{n'N'}, E - \hbar\omega, k_\parallel) \Delta^+_\omega(r'_{n'N'}) \langle r'_{n'N'} \mid \psi^{\tau'}_{k_\parallel E}\rangle. \end{aligned} \tag{25b}$$

The atomic summation $\sum_{N,n}$ is done over one unit cell of the Nth monolayer and $J_{\mathrm{cell},N}$ is the number of unit cells within the Nth monolayer. With the aid of the spin-density matrix ρ the spin-averaged photocurrent $\bar{I}$, the photoelectron spin polarization $\boldsymbol{P}$ and spin-resolved photocurrent I_τ can be expressed as $\bar{I} = \mathrm{Tr}\,\rho$, $\boldsymbol{P} = \mathrm{Tr}[\sigma\rho]/\bar{I}$, $I_\tau = (1 + \tau \boldsymbol{P}\cdot\boldsymbol{n})\bar{I}/2$, respectively, where the unit vector $\boldsymbol{n}$ defines the direction of the spin analysis. The retarded GF $G^{\mathrm{tot}}(\boldsymbol{r}_{nN}, \boldsymbol{r}'_{n'N'}, E-\hbar\omega, \boldsymbol{k}_\parallel)$ as usual describes all the scattering paths for the hole going from $\boldsymbol{r}_{nN}$ at the nth atom of the Nth monolayer to $\boldsymbol{r}'_{n'N'}$. The interaction of the electron with a monochromatic electromagnetic field given by $\boldsymbol{a}(\boldsymbol{r})\mathrm{e}^{\mathrm{i}\omega t}$, where $\boldsymbol{a}(\boldsymbol{r})$ is the spatial part of the magnetic vector potential, is given as

$$\Delta_\omega(\boldsymbol{r}) = \hat{\boldsymbol{\alpha}}\cdot\boldsymbol{a}(\boldsymbol{r})\mathrm{e}^{\mathrm{i}\omega t}. \tag{26}$$

We note that $\boldsymbol{a}(\boldsymbol{r})$ should actually incorporate the optical response of the solid (especially in the surface region). In most applications $\boldsymbol{a}(\boldsymbol{r})$ has so far been approximated by $\boldsymbol{a}_0\mathrm{e}^{\mathrm{i}q\boldsymbol{r}}$, where $\boldsymbol{a}_0$ is the amplitude in the vacuum region, or simply by $\boldsymbol{a}_0$, which corresponds to the electric dipole approximation. The final state $\langle \boldsymbol{r} \mid \psi^\tau_{\boldsymbol{k}_\parallel E}\rangle$ is related to a so-called low-energy electron diffraction (LEED) state by the time-inversion operation $\hat{K}$ with the appropriate phases for the different spin states $\tau = \pm 1/2$:

$$\begin{aligned}\langle \boldsymbol{r} \mid \psi^{\pm 1/2}_{\boldsymbol{k}_\parallel E}\rangle &= \langle \boldsymbol{r}\mid \hat{K}\,G^{\mathrm{tot}}(\boldsymbol{r}_{nN}, \boldsymbol{r}'_{n'N'}, E, \boldsymbol{k}_\parallel)\hat{K}^+ \mid \phi^{\pm 1/2}_{\boldsymbol{k}_\parallel E}\rangle \\ &= \mp\hat{K}\langle \boldsymbol{r}\mid \psi^{\mp 1/2}_{-\boldsymbol{k}_\parallel E}\rangle^{\mathrm{LEED}} \equiv \hat{T}\langle \boldsymbol{r}\mid \psi^{\mp 1/2}_{-\boldsymbol{k}_\parallel E}\rangle^{\mathrm{LEED}}\end{aligned} \tag{27}$$

where $\langle \boldsymbol{r} \mid \phi^s_{\boldsymbol{k}_\parallel E}\rangle$ describes the free-electron wavefunction at the plane of observation, which is parallel to the surface.

Using our Green function results (section 3), we now develop equation (25) into a more explicit form, which makes its physical content more visible and which can be employed in numerical computations. The final state can be written at the nth atom of the Nth monolayer in the spherical-wave representation as

$$\langle \boldsymbol{r}_{nN} \mid \psi^\tau_{\boldsymbol{k}_\parallel E}\rangle = \frac{1}{S_N^{1/2}}\hat{T}\sum_{\kappa\mu} A^{N\tau}_{\kappa\mu n}\langle \boldsymbol{r}_{nN}\mid \psi^{\mathrm{R}}_{\kappa\mu}\rangle \tag{28}$$

where as before $|\psi^{\mathrm{R}}_{\kappa\mu}\rangle$ means the Dirac bispinor regular at the origin. The amplitudes of this expansion $A^{N\tau}_{\kappa\mu n}$ are calculated by a layer T-matrix combination scheme starting from the free state $\langle \boldsymbol{r} \mid \phi^s_{\boldsymbol{k}_\parallel E}\rangle$ at the plane of observation and going up to the given Nth monolayer. Substituting equation (15) for the GF (with one of the layer labels N replaced by N') and equation (28) for the final state into equation (25*b*), we get

$$\begin{aligned}\tilde{\rho}_{\tau\tau'}(\boldsymbol{k}_\parallel, E) = -\frac{1}{\pi}k\sum_{N,n}\sum_{N',n'}\int \mathrm{d}\boldsymbol{r}_{nN}\int \mathrm{d}\boldsymbol{r}'_{n'N'}\frac{J_{\mathrm{cell},N}}{S_N^{1/2}}\frac{J_{\mathrm{cell},N'}}{S_{N'}^{1/2}} \\ \times\sum_{\kappa\mu}\{A^{N\tau *}_{\kappa\mu n}\langle\psi^{\mathrm{R}}_{\kappa\mu}\mid \boldsymbol{r}_{nN}\rangle\hat{T}^+_{\Leftarrow}\Delta_\omega(\boldsymbol{r}_{nN})\}_E \\ \times\left\{G_{\mathrm{a}}(\boldsymbol{r}_{nN}, \boldsymbol{r}'_{n'N'})\delta_{nN,n'N'} + \sum_{\substack{\kappa\mu\\ \kappa'\mu'}} U^{NN'}_{\substack{\kappa\mu n\\ \kappa'\mu' n'}}\langle \boldsymbol{r}_{nN}\mid\psi^{\mathrm{R}}_{\kappa\mu}\rangle\langle\psi^{\mathrm{R}}_{\kappa'\mu'}\mid \boldsymbol{r}'_{n'N'}\rangle\right\}_{E-\hbar\omega} \\ \times\left\{\Delta^+_\omega(\boldsymbol{r}'_{n'N'})\hat{T}_{\Rightarrow}\sum_{\kappa\mu}A^{N'\tau'}_{\kappa\mu n'}\langle \boldsymbol{r}_{n'N'}\mid\psi^{\mathrm{R}}_{\kappa\mu}\rangle\right\}_E\end{aligned} \tag{29}$$

where $G_a(r_{nN}, r'_{nN})$ is the atomic-like GF (equation (16)).

Eventually we obtain

$$\tilde{\rho}_{\tau\tau'}(k_\parallel, E) = -\frac{1}{\pi}k\Big(- \mathrm{i}k \sum_{N,n} \frac{1}{S_{\mathrm{cell},N}} \sum_{\kappa\mu} \sum_{\kappa'\mu'} \int \mathrm{d}r_{nN} \int \mathrm{d}r'_{n'N'} (\{A^{N\tau *}_{\kappa\mu n} \langle \psi^{\mathrm{R}}_{\kappa\mu} | \hat{T}^{+}_{\Leftarrow} \Delta^{+}_{\omega} | r_{nN} \rangle\}_E$$

$$\times \langle r_{nN} | G_a | r'_{nN} \rangle_{E-\hbar\omega} \{ \langle r'_{nN} | \Delta_\omega \hat{T}_{\Rightarrow} | \psi^{\mathrm{R}}_{\kappa'\mu'} \rangle A^{N\tau'}_{\kappa'\mu' n} \}_E)$$

$$+ \sum_{\substack{N,n \\ N',n'}} \frac{1}{S^{1/2}_{\mathrm{cell},N}} \frac{1}{S^{1/2}_{\mathrm{cell},N'}} \sum_{\kappa\mu} \sum_{\kappa'\mu'} D^{N\tau *}_{\kappa\mu n} U^{NN'}_{\substack{\kappa\mu n \\ \kappa'\mu' n'}} D^{N'\tau'}_{\kappa'\mu' n'} \Big) \tag{30}$$

where

$$D^{N\tau}_{\kappa\mu n} = \sum_{\kappa'\mu'} \int \mathrm{d}r_{nN} \langle \psi^{\mathrm{R}}_{\kappa\mu} \mid r_{nN} \rangle_{E-\hbar\omega} \{ \langle r_{nN} | \Delta_\omega \hat{T}_{\Rightarrow} | \psi^{\mathrm{R}}_{\kappa\mu} \rangle A^{N\tau}_{\kappa'\mu' n} \}_E$$

and $S_{\mathrm{cell},N}$ denotes the unit cell area of the Nth monolayer. The photocurrent thus manifestly is a sum of contributions from the different atoms in the unit cell of a layer (index n) and from the individual layers (index N) (the number of which is determined by the decay of the final state). For each pair (N, n), the first term in equation (30) (involving the atomic 'lower-state' atomic Green function G_a) represents a contribution from the (hypothetically) isolated single crystal atom and the second one (characterized by the 'lower-state' boundary correction coefficients U) gives contributions due to intra- and interlayer multiple scattering of the lower-state wave field. We note that the above formulation allows for inelastic scattering within the core region. We do not elaborate here on the integrals ('matrix elements') in equation (30), since these have the same structure as in related work, where they have already been discussed in detail (Ackermann and Feder 1985a, Braun *et al* 1985, 1987, Ginatempo *et al* 1989).

5. Conclusion

We have developed a fully relativistic Green function formalism to connect different atomic layers (with several atoms per unit cell) of a semi-infinite crystalline system—including adsorbates and thin films—which may be ferromagnetic. The underlying single-particle effective potential can be complex and need not be spherically symmetric inside atomic spheres. From the 'same-layer' special case of this Green function we derived layer-, spin- and symmetry-resolved densities of states. The general form was employed to treat—in a dynamical way—the 'lower-state' part in a relativistic one-step model photoemission formalism. This allows one to include the hole lifetime from the start. As a by-product we obtain the complex bulk band structure via diagonalization of the matrix, which transfers the total wave field across a bulk layer. In deriving our Green function, we used in an intermediate step an 'empty-layer Green function' relating to the semi-infinite system with the 'current' two layers removed. An alternative approach consists of removing only one layer, the source layer of the Green function, calculating the corresponding empty-layer Green function for z at the layer boundary and then propagating it through the adjacent atomic layers up to a second layer (which is not empty, in contrast to our above formulation). This empty-layer Green function is then used in a Dyson equation to obtain the actual GF.

A third approach (which is a modification of the previous one) employs the single empty-layer Green function to obtain the actual Green function at the boundary of the source layer. The actual GF is then propagated to the second layer and re-expanded with respect to spherical solutions within this layer. These seemingly different approaches are of course mathematically equivalent. It is a question of computational convenience which—if any—might actually be preferable over the others. Numerical work employing the layer density of states and photoemission expressions presented here is currently in progress.

The present layer Green function is also a key element for further theoretical developments like the practical inclusion of higher-order Keldysh diagrams into photoemission formalisms and a realistic treatment of optical and magneto-optical processes in surface and thin-film systems.

References

Ackermann B 1985 *PhD Thesis* Universität Duisburg

Ackermann B and Feder R 1985a *J. Phys. C: Solid State Phys.* **18** 1093

—— 1985b *Solid State Commun.* **54** 1077

Braun J, Nolting W and Borstel G 1991 *Surf. Sci.* **251/252** 22

Braun J, Thörner G and Borstel G 1985 *Phys. Status Solidi* b **130** 643

—— 1987 *Phys. Status Solidi* b **144** 609

Caroli C, Lederer-Rozenblatt D, Roulet B and Saint-James D 1973 *Phys. Rev.* B **8** 4552

Feder R 1985 (ed) *Polarized Electrons in Surface Physics* (Singapore: World Scientific)

Ginatempo B, Durham P J and Gyorffy B I 1989 *J. Phys.: Condens. Matter* **1** 6483

Ginatempo B, Durham P J, Gyorffy B I and Temmerman W M 1985 *Phys. Rev. Lett.* **54** 1581

Ginatempo B and Gyorffy B I 1990 *J. Phys.: Condens. Matter* **2** 5233

Gollisch H and Feder R 1990 *Solid State Commun.* **76** 237

Heinzmann U 1990 *Photoemission and Absorption Spectroscopy of Solids and Interfaces with Synchrotron Radiation* ed M Campagna and R Rosei (Amsterdam: North-Holland)

Hillebrecht F U, Jungblut R and Kisker E 1990 Phys. Rev. Lett. 65 2450

Hora R and Scheffler M 1984 *Phys. Rev.* B **29** 629

Keldysh L V 1965 *Sov. Phys.–JETP* **20** 1018

Kessler B, Eyers A, Horn K, Müller N, Schmiedeskamp B, Schönhense G and Heinzmann U 1987 *Phys. Rev. Lett.* **59** 331

Kirschner J 1985 *Polarized Electrons at Surfaces (Springer Tracts in Modern Physics 106)* (Heidelberg: Springer)

Kisker E 1987 *Metallic Magnetism* ed H Capellmann (Heidelberg: Springer)

Koenig U, Hoermandinger G, Weinberger P, Ginatempo B and Boring A M 1991 *Physica* B **172** 125

Pendry J B 1976 *Surf. Sci.* **57** 679

Schneider C M, Garbe J, Bethke K and Kirschner J 1989 *Phys. Rev.* B **39** 1031

Schneider C M, Hammond M S, Schuster P, Cebollada A, Miranda R and Kirschner J 1991 *Phys. Rev.* B **44** 12 066

Stoppmanns P, Heidemann B, Irmer N, Müller N, Vogt B, Schmiedeskamp B, Heinzmann U, Tamura E and Feder R 1991 *Phys. Rev. Lett.* **66** 2645

Tamura E 1992a *Phys. Rev.* B **45** 3271

—— 1992b *Applications of Multiple Scattering Theory to Materials Science* ed W H Butler, P H Dederichs, A Gonis and R L Weaver (Pittsburgh: Materials Research Society)

Tamura E and Feder R 1989 *Solid State Commun.* **70** 205

—— 1991a *Solid State Commun.* **79** 989

—— 1991b *Europhys. Lett.* **16** 695

Tamura E, Piepke W and Feder R 1987 *Phys. Rev. Lett.* **59** 934

Tamura E, Feder R, Vogt B, Schmiedeskamp B and Heinzmann U 1989 *Z. Phys.* B **77** 129