DOUBLE-ELECTRON PHOTOEMISSION FROM SURFACES

Theory and Application to Cu(001)

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Abstract A general theory of double-electron photoemission (DPE) from solid surfaces is sketched. Guided by the one-step model of single-electron photoemission (SPE), DPE can be related closely to SPE if the electronic correlation is approximated by a local but wave-vector-dependent effective one-particle potential. Numerical results for DPE from Cu(001) confirm the expected manifestation of the electronic correlation, in particular the correlation hole.

Keywords: double-electron photoemission, electronic correlation, Cu(001)

1. PROLOGUE

Double-electron photoemission (DPE) from solids and surfaces is perfectly suited for studying the dynamics of correlated electrons, as has been demonstrated experimentally by Herrmann et al., 1998. Detecting simultaneously two electrons after the absorption of one photon, this process is one of the few known that in principle cannot be described within the one-particle (OP) picture because electronic correlation substantially influences the characteristics of the doubly excited states. The failure of the OP picture becomes evident if for instance the initial-state and the final-state two-electron wave functions are approximated by products of orthogonal one-electron wave functions: the transition-matrix elements for double photoionization vanish (Berakdar, 1998). Therefore, quanti-

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ties used for the theoretical formulation of the problem (wave functions, propagators, etc.) have to be of at least two-particle (TP) nature.

As is well-known, scattering of the electrons in the solid affects considerably the intensities of single-electron photoemission (SPE). Based on the well-established one-step model in the sudden approximation (see e. g. Kevan, 1992), available computer codes successfully treat SPE, in particular from valence bands of transition and noble metals. Therefore, it appears desirable to have an extension of the SPE theory to DPE, in order to approach experiment. In this Paper, we sketch a theory of DPE from solid surfaces, give a formulation of it in close connection to SPE, and present numerical results for photoemission from Cu(001).

2. DOUBLE-ELECTRON PHOTOEMISSION

Theoretical descriptions of SPE are usually based on the work of Feibelman and Eastman, 1974, who within the framework of non-equilibrium Green functions had derived the one-step model of photoemission (see e. g. Schattke, 1997). In the sudden approximation, i. e., neglecting the interaction of the photo-electron with the hole and the remaining electrons (Hedin et al., 1998), the photocurrent is given by

$$J(\boldsymbol{k}) = -\frac{1}{\pi}\left\langle \psi \middle| \Delta \,\mathrm{Im}\, g^{\mathrm{r}}(E-\omega)\, \Delta^{\dagger} \middle| \psi \right\rangle, \tag{1}$$

where $\boldsymbol{k}$ is the wave-vector of the photo-electron at the detector (Fig. 1). The kinetic energy E and the surface-parallel wave-vector $\boldsymbol{k}^{\parallel}$ determine the surface-normal component of $\boldsymbol{k}$ by $k^{\perp} = \sqrt{2E - (\boldsymbol{k}^{\parallel})^2}$ (We use atomic units, $\hbar = m = e = 1$). Reading the above equation from the right, the SPE process can be portrayed as follows. In vacuum, an incoming electron with energy E is described by a plane wave $|\boldsymbol{k}\rangle$. The latter is propagated into the solid by the advanced Green function g^{a},

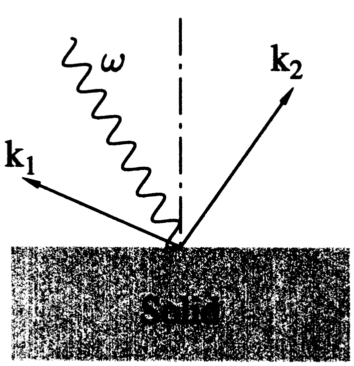

Figure 1 Sketch of the experimental set-up. A photon with energy ω impinges onto the solid (wavy line). In single-electron photoemission, one electron with momentum $\boldsymbol{k}_1$ is detected. In double-electron photoemission, two outgoing electrons with momenta $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$ are detected in coincidence.

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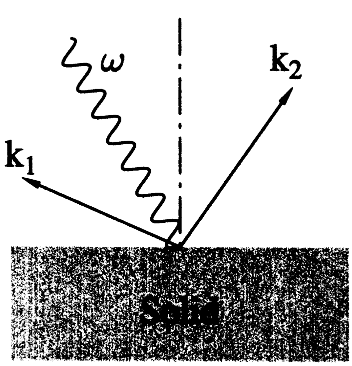

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leading to the wave function $|\psi\rangle = g^{\mathrm{a}}\,|\boldsymbol{k}\rangle$ (We use lowercase letters for OP wave and Green functions). Note that the time-reversed $|\psi\rangle$ is used in the theory of low-energy electron diffraction (LEED). Subsequently, $|\psi\rangle$ is de-excited by the dipole operator $\Delta^{\dagger}$ into occupied states with energy $E-\omega$, the latter being described by the imaginary part of the retarded Green function, $\operatorname{Im} g^{\mathrm{r}}(E-\omega)$. Eventually, the electron is excited by Δ and propagated back to the detector. The SPE process is visualized by the (lowest-order) Feynman diagram shown in Fig. 2. The sudden approximation suggests a 'natural' separation into an 'active' electron, i.e., the outgoing photo-electron, and 'passive' electrons, the remaining ones. As we shall show now, the above ideas can directly be applied to DPE.

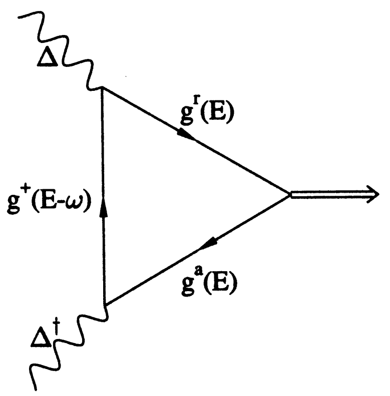

Figure 2 Diagrammatic representation of the photocurrent expression eq. (1). Wavy lines represent photons, straight lines with arrows correspond to retarded and advanced electron Green functions g^{r} and g^{a} at the final and initial state energies E and $E-\omega$. The double line symbolizes the emitted (and detected) photo-electron.

In DPE, one detects two outgoing electrons in coincidence upon an incoming photon. The detection of both photo-electrons allows for the simultaneous determination of their kinetic energies E_1 and E_2 as well as $\boldsymbol{k}_1^{\parallel}$ and $\boldsymbol{k}_2^{\parallel}$ (Fig. 1). The experiment can be characterized by the sums of the kinetic energies, $E = E_1 + E_2$, and of $\boldsymbol{K}^{\parallel} = \boldsymbol{k}_1^{\parallel} + \boldsymbol{k}_2^{\parallel}$. Since OP momenta are in general not 'good' quantum numbers in DPE (due to the electron-electron interaction), conservation of $\boldsymbol{k}_{\parallel}$ (modulo reciprocal surface-lattice vectors) in SPE translates into conservation of $\boldsymbol{K}^{\parallel}$ in DPE (Berakdar, 1998).

As in the sudden approximation of SPE, we separate the electrons into the 'active' photo-electrons and the 'passive' ground-state electrons. First note that the electron-electron interaction between all electrons is taken into account within density-functional theory. In the spirit of the sudden approximation, the interaction of the 'active' pair of electrons with the 'passive' electrons is neglected. However, the (screened) Coulomb interaction within the 'active' pair has explicitly to be taken into account. Otherwise the simultaneous excitation of two correlated electrons would not be possible. By this means, three-particle and higher-order terms are neglected.

In analogy to the one-step model of SPE, the DPE current can be expressed as in eq. (1), but with OP quantities replaced by TP ones (We use uppercase letters for TP wave and Green functions). For example, $|\Psi\rangle$ becomes the time-reversed LEED state of two correlated electrons, e. g., it has the boundary conditions of plane waves $|\boldsymbol{k}_1\rangle$ and $|\boldsymbol{k}_2\rangle$ taken at the detector positions. The latter is propagated by the advanced TP-Green function G^{a} from inside the solid towards the detectors, $|\Psi\rangle = G^{\mathrm{a}}\,|\boldsymbol{k}_1, \boldsymbol{k}_2\rangle$. Further, $\Delta = \Delta_1 + \Delta_2$ is the TP dipole operator, i. e., the sum of two OP dipole operators, and $-\mathrm{Im}\, G^{\mathrm{r}}(E-\omega)/\pi$ is the non-local density of TP states.

Assuming infinite TP life-times, $\mathrm{Im}\, G^{\mathrm{r}}(E)$ can be written in terms of TP states $|\Phi_i\rangle$ with energies E_i and further quantum numbers comprised in the compound index i, $-\mathrm{Im}\, G^{\mathrm{r}}(E) = \pi \sum_i |\Phi_i\rangle\, \delta(E - E_i)\, \langle\Phi_i|$. Using this expression, the DPE current is given by Fermi's 'golden rule',

$$J(\boldsymbol{k}_1, \boldsymbol{k}_2) = \sum_i |\, \langle\Psi|\Delta|\Phi_i\rangle\, |^2\, \delta(E - \omega - E_i). \tag{2}$$

In order to evaluate the above DPE current formulae, eqs. (1) and (2), ways have to be found to calculate the TP-Green function or the TP states involved. We shall address this problem in the following section.

3. TREATMENT OF TWO-ELECTRON STATES

The Hamiltonian for two electrons inside a semi-infinite solid is given by $H = K + U + W$, i. e., the sum of kinetic energy K, electron-electron interaction U, and the solid's potential W. Due to the simultaneous occurrence of the many-body (U) and the scattering (W) problem, the sequence of treating them appears to be crucial.

Remembering that DPE is due to the Coulomb interaction, U is assigned to the reference Hamiltonian $H_{\mathrm{int}} = K + U$, thus keeping W as a perturbation. The TP state $|\Phi\rangle$ could therefore be calculated as follows. From the un-correlated state $|\Phi_0\rangle$, an eigenfunction of K, the correlated state $|\Phi_{\mathrm{int}}\rangle$, an eigenfunction of H_{int}, can be obtained by $|\Phi_{\mathrm{int}}\rangle = (1 + G_{\mathrm{int}} U)\,|\Phi_0\rangle$ with $G_{\mathrm{int}} = (E - H_{\mathrm{int}})^{-1}$. Eventually, multiple-scattering in the solid leads to $|\Phi\rangle = (1 + G_{\mathrm{int}} T)\,|\Phi_{\mathrm{int}}\rangle$, with T denoting the transition operator (see e. g. Gonis, 1992). The above scheme is general and applicable for both initial and final TP states of DPE. However, as the central problem remains that due to G_{int} the 'internal' motion of the 'active' electron pair cannot be separated from its 'external' motion.

As an alternative to the above procedure, we consider an approach for the calculation of the final TP state $|\Psi\rangle$, which uses OP scattering states as reference states. These states are well-known from LEED and SPE theories and can be calculated for example by multiple-scattering methods. The TP final state $|\Psi\rangle$ is expressed in terms of OP final states $|\psi_1\rangle$ and $|\psi_2\rangle$ as $|\Psi\rangle = (1 + G_{\text{int}}U)\,|\psi_1, \psi_2\rangle$. According to Thomas-Fermi theory, the screened Coulomb interaction U with screening length λ is given by

$$U = \frac{\exp(-|\boldsymbol{r}_1 - \boldsymbol{r}_2|/\lambda)}{|\boldsymbol{r}_1 - \boldsymbol{r}_2|} = \frac{Z_1}{r_1} + \frac{Z_2}{r_2}, \tag{3}$$

where we have introduced $Z_j = \exp(-a_j r_j/2\lambda)/a_j$ and $a_j = 2|\boldsymbol{r}_1 - \boldsymbol{r}_2|/r_j$, $j = 1, 2$. Thus, U can be partitioned in $W + U$ as

$$\underbrace{w(\boldsymbol{r}_1) + w(\boldsymbol{r}_2)}_{=W} + U(\boldsymbol{r}_1 - \boldsymbol{r}_2) = \underbrace{w(\boldsymbol{r}_1) + \frac{Z_1}{r_1}}_{=\widetilde{w}_1} + \underbrace{w(\boldsymbol{r}_2) + \frac{Z_2}{r_2}}_{=\widetilde{w}_2}, \tag{4}$$

with OP potentials $w(\boldsymbol{r}_j)$. Of course, eq. (4) is exact and nothing is gained by introducing the effective non-local potentials $\widetilde{w}(\boldsymbol{r}_1, \boldsymbol{r}_2)$. However, by approximating a_j as $2|\boldsymbol{k}_1 - \boldsymbol{k}_2|/k_j$, the Coulomb interaction is forced to be local and to depend on the wave-vectors $\boldsymbol{k}_j$. In this way, the electron-electron interaction can be included in the OP potential w which so becomes dependent on $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$, i.e., $w(\boldsymbol{r}_1) \to \widetilde{w}(\boldsymbol{r}_1; \boldsymbol{k}_1, \boldsymbol{k}_2)$ and analogously for $w(\boldsymbol{r}_2)$. The interpretation of DPE is now as follows (Fig. 3): one 'active' electron absorbs the photon and thus changes the effective potential w into $\widetilde{w}$. The latter is 'felt' by the other 'active' electron and results in a 'shake-up' of this electron.

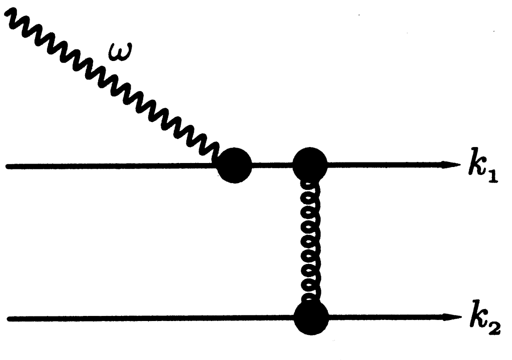

Figure 3 Sketch of correlated emission of two electrons. An electron is scattered at a photon with energy ω (wavy line) and emitted with wave vector $\boldsymbol{k}_1$. Due to the change of the effective potential (spiral), the second electron is subject to a 'shake-up' process and emitted with wave vector $\boldsymbol{k}_2$.

The predominant effect of this 'local' approximation on w is illustrated in Fig. 4. The reduction of the potential depth can be regarded as a reduction or screening of the (positive) core charge. This would lead to a repulsion of the electrons from the site under consideration. In other

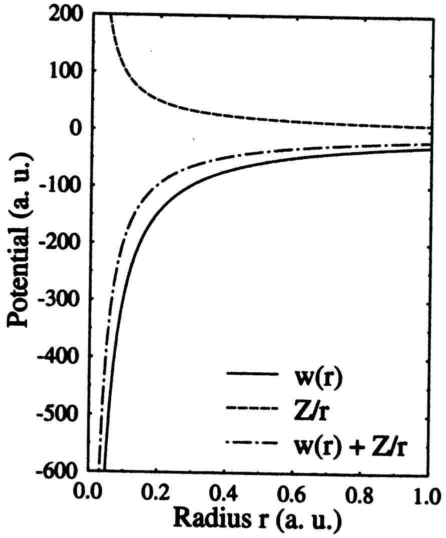

Figure 4 Effect of the 'local' approximation on the one-particle (OP) potential. The OP potential $w(r) = 29/r$ (solid line) is changed by the approximated electron-electron interaction Z/r with screening length $\lambda = 1$ and $a = 0.1$ according to eq. (4) (dashed). The resulting potential $\widetilde{w} = w + Z/r$ is given by the dashed-dotted line.

words, one would observe a correlation hole. Further, if $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$ are close the change in $\widetilde{w}$ is strong, whereas if they are remote the change is weak. In summary, the above approximation which has given good results in low-energy (e, 2e) spectroscopy (see e.g. Feder et al., 1998, and Gollisch et al., 1999) is expected to describe at least qualitatively the electronic correlation in the TP final state of DPE, in particular the correlation hole. We now apply the 'local' approximation to DPE from valence bands.

4. VALENCE-BAND DOUBLE-ELECTRON PHOTOEMISSION

The ground state of metals with dispersive valence bands can be well described by independent quasi-electrons. Therefore, it appears reasonable to neglect explicitly the correlation in the TP initial state, e.g., the OP initial states being eigenfunctions of the ground-state Hamiltonian $K + W$. The TP final state is taken as an anti-symmetrized product of OP final states $\left|\widetilde{\psi}_j\right\rangle = \widetilde{g}_j^{a} \left|\boldsymbol{k}_j\right\rangle$, i.e., $\left|\widetilde{\Psi}\right\rangle = \mathcal{A}\left(\left|\widetilde{\psi}_1\right\rangle, \left|\widetilde{\psi}_2\right\rangle\right)$ with $\mathcal{A}$ denoting anti-symmetrization and the tilde indicating eigenfunctions of $K + \widetilde{W}$. Thus, correlation is treated explicitly only in the final state. In this approximation, the photocurrent can be expressed as difference of a direct term (I_{d}) and an exchange term (I_{e}),

$$J(\boldsymbol{k}_1, \boldsymbol{k}_2) \sim \frac{1}{\pi^2} \int_{E_{\min}}^{E_{\mathrm{F}}} \left[I_{\mathrm{d}}(E') - I_{\mathrm{e}}(E')\right] \mathrm{d}E', \tag{5}$$

with $E_{\mathrm{min}} = E - \omega - E_{\mathrm{F}}$ being the lowest occupied OP energy (for details see Fominykh et al., 2000). The direct contribution I_{d} consists of four terms,

$$\begin{aligned} I_{\mathrm{d}}(E') &= \left\langle \tilde{\psi}_1 \middle| \Delta \mathrm{Im}\, g^{\mathrm{r}}(E') \Delta^{\dagger} \middle| \tilde{\psi}_1 \right\rangle \left\langle \tilde{\psi}_2 \middle| \mathrm{Im}\, g^{\mathrm{r}}(E'') \middle| \tilde{\psi}_2 \right\rangle \\ &+ \left\langle \tilde{\psi}_1 \middle| \Delta \mathrm{Im}\, g^{\mathrm{r}}(E') \middle| \tilde{\psi}_1 \right\rangle \left\langle \tilde{\psi}_2 \middle| \mathrm{Im}\, g^{\mathrm{r}}(E'') \Delta^{\dagger} \middle| \tilde{\psi}_2 \right\rangle \\ &+ \left\langle \tilde{\psi}_1 \middle| \mathrm{Im}\, g^{\mathrm{r}}(E') \Delta^{\dagger} \middle| \tilde{\psi}_1 \right\rangle \left\langle \tilde{\psi}_2 \middle| \Delta \mathrm{Im}\, g^{\mathrm{r}}(E'') \middle| \tilde{\psi}_2 \right\rangle \\ &+ \left\langle \tilde{\psi}_1 \middle| \mathrm{Im}\, g^{\mathrm{r}}(E') \middle| \tilde{\psi}_1 \right\rangle \left\langle \tilde{\psi}_2 \middle| \Delta \mathrm{Im}\, g^{\mathrm{r}}(E'') \Delta^{\dagger} \middle| \tilde{\psi}_2 \right\rangle, \end{aligned} \tag{6}$$

with $E'' = E - \omega - E'$. For the exchange contribution I_{e}, one has to replace $\left|\tilde{\psi}_1\right\rangle \left\langle \tilde{\psi}_2\right|$ by $\left|\tilde{\psi}_2\right\rangle \left\langle \tilde{\psi}_1\right|$. Again, a 'golden rule' form can be derived,

$$\begin{aligned} J(\boldsymbol{k}_1, \boldsymbol{k}_2) &\sim \sum_{kl}^{\mathrm{occ}} | \overbrace{M_{kl}^{(12)} + M_{kl}^{(21)}}^{\text{direct}} - \overbrace{M_{lk}^{(12)} - M_{lk}^{(21)}}^{\text{exchange}} |^2 \\ &\quad \times \delta(E - \omega - \epsilon_k - \epsilon_l), \end{aligned} \tag{7}$$

where the TP-transition-matrix elements $M_{kl}^{(ij)}$ are products of usual OP-transition-matrix elements and overlap integrals, $M_{kl}^{(ij)} = \left\langle \tilde{\psi}_i \middle| \Delta \middle| \phi_k \right\rangle \left\langle \tilde{\psi}_j \middle| \phi_l \right\rangle$. The overlap is non-zero because $\left|\tilde{\psi}_j\right\rangle$ and $|\phi_l\rangle$ are eigenfunctions of different Hamiltonians, e.g., the former (latter) of $K + \widetilde{W}$ $(K + W)$.

Considering equations (5) and (6), it is straightforward to enhance existing SPE computer codes in order to treat DPE. These codes already calculate matrix elements as in eq. (6), as is obvious from comparison with eq. (1).

5. NUMERICAL RESULTS FOR Cu(001)

We have implemented the above theory and its approximations into our spin-polarized relativistic layer-Korringa-Kohn-Rostoker (LKKR) code for electron spectroscopies (Henk et al., 1999). As a first application, we performed DPE calculations for the prototypical system Cu(001) in order to reveal qualitative trends. The ground-state potentials have been calculated self-consistently with the scalar-relativistic linearized muffin-tin orbital (LMTO) method. Life-time broadening of the spectra was simulated by use of a complex optical potential. The photocurrent due to emission from the first 20 outermost layers was calculated from the layer-resolved Green function according to eqs. (5) and (6). As usual, extensive convergence tests on, e. g., the maximum angular momentum, number of reciprocal lattice vectors, and accuracy of the energy integration in eq. (5) were carried out.

As a typical example, we compare in Fig. 5 the angular intensity distribution of SPE (l. h. s.) with that of DPE (r. h. s.) for incoming s-polarized light and fixed kinetic energy of the photo-electrons (E_{kin} = 9 eV). In SPE, the detector position is varied in the whole hemisphere, whereas in DPE the position of one detector is also varied while the other is fixed. Further, the photon energy is 31 eV in DPE, but only 15.5 eV in SPE. Therefore, the initial-state energy in SPE is in the centre of the energy-integration range in DPE (cf. eq. (5)).

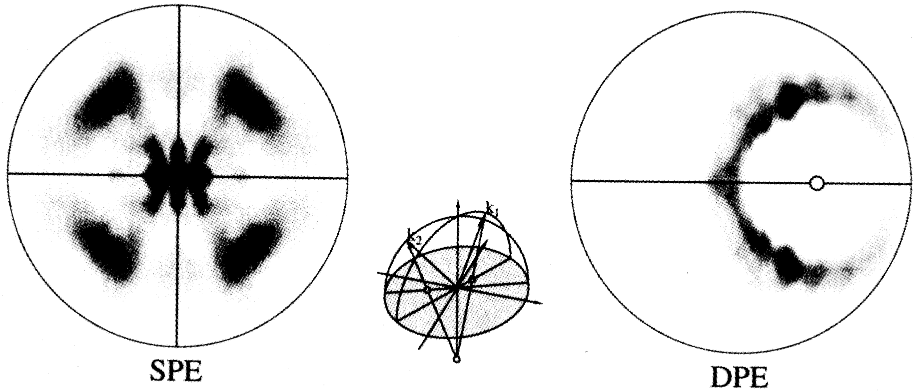

Figure 5 Angular distribution of the photoemission intensity from Cu(001) in single-electron (SPE, l. h. s.) and double-electron (DPE, r. h. s.) photoemission. The kinetic energy of the photo-electrons is 9 eV, the photon energy of the s-polarized light 15.5 eV in SPE and 31 eV in DPE. In the right inset, the small circle indicates the fixed position of one of the detectors (polar angle $\vartheta = 40°$). Low (high) intensities correspond to light (dark) gray scale in the stereo-graphic projection (see middle inset). Horizontal and vertical lines emphasize the symmetries of the angular distributions.

In SPE, the point group of the surface $4mm$ is reduced to $2mm$ in the angular distribution (as indicated by the horizontal and vertical lines in Fig. 5) because the electric-field vector of the incident light (which lies in a mirror plane of the surface) is not invariant under operations C_4 and C_4^{-1}. In DPE, the group $2mm$ is reduced further to m (horizontal line in Fig. 5) by fixing the position of one of the detectors (as is indicated by the small circle, $\vartheta = 40°$, thus fixing say $\boldsymbol{k}_2$).

The electronic correlation has a profound effect on the DPE intensity. For the screening length λ chosen as 1.0 a. u., we observe a drastic reduction of the intensity at angles near the fixed detector position, i. e., if $\boldsymbol{k}_1$

and $\boldsymbol{k}_2$ are close. This void is surrounded by a circular intensity distribution. At higher angles, the intensity is again almost zero. This angular distribution directly shows the signatures of the electronic correlation, e. g., the correlation hole.

In order to corroborate the origin of the correlation hole, we performed calculations for different values of the screening length λ, see Fig. 6. Again, we observe a correlation hole, the size of which depends on the value of λ. A small λ yields a small void around the the fixed detector, whereas a large λ gives rise to an extended intensity void, as expected from eqs. (3) and (4).

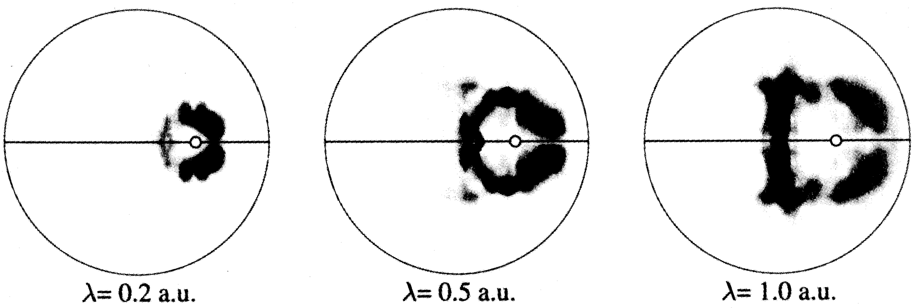

Figure 6 Effect of the screening length λ on the angular distribution in DPE from Cu(001). S-polarized light with 45 eV photon energy impinges onto the surface, the kinetic energy is fixed at 15 eV. The stereo-graphic images show angular distributions for $\lambda = 0.2$ a. u. (left), 0.5 a. u. (middle), and 1.0 a. u. (right). Vertical lines emphasize the mirror symmetry of the angular distributions.

In the calculations for Figs. 5 and 6, the kinetic energy E_{kin} of the photo-electrons has changed and, therefore, different electronic states are involved in the photoemission process. In Fig. 5 we found an almost perfect circular intensity shape, whereas in Fig. 6 the intensity appears to be more square-like, in particular for $\lambda = 1.0$ a. u. This directly proves the profound effect of the electronic structure on the DPE intensities.

6. EPILOGUE

We have outlined a general theory of double-electron photoemission (DPE) from solid surfaces. A local but wave-vector-dependent approximation of the Coulomb interaction between the photo-electrons gives a close connection of DPE to the well-established one-step model of single-electron photoemission (SPE). Therefore, DPE can be treated numerically with enhanced SPE computer codes. First numerical results for DPE from

Cu(001) reveal the fingerprints of the electronic correlation, in particular the correlation hole, and confirm the expected qualitative trends.

In view of magnetic dichroism in DPE, our scheme is currently extended to magnetic systems. Another approach to be investigated is the explicit treatment of the screened Coulomb interaction (Gollisch et al., 1992).

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