

Bulk- and surface-state two-electron photoemission from Cu(1 1 1)

N. Fominykh^a, J. Berakdar^{b,*}

^a Max-Planck Institut für Mikrostrukturphysik, Weinberg 2, 06120 Halle, Germany

^b Institute of Physics, Martin-Luther-University Halle-Wittenberg, Heinrich-Damerow-Str. 4, Nanotechnikum Weinberg, 06120 Halle, Germany

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Abstract

We study theoretically the one-photon two-electron emission from the Cu(1 1 1) surface. Tuning the energy to either the surface state or to the d-band energy regions this sample provides a paradigm example of the two-particle emission from a (quasi)-two-dimensional and a three-dimensional system, respectively. The independent electron–gas behavior of the surface-state electrons results in a characteristic angular pattern. We inspect the dependence of this angular shape on the strength of the electron–electron interaction. Furthermore, it is shown that the two-particle energy correlation behaves differently for bulk and surface states.

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1. Introduction

Over the past few years impressive experimental advances have been accomplished in the momentum-resolved detection of a correlated electron pair emitted from solid surfaces upon a single photon absorption [1,2]. A prime goal of these double photoemission (DPE) experiments is to uncover and to elucidate the influence of electron interactions in solids, for a DPE description must go beyond independent-particle models [3].

For an inhomogeneous many-body system such as a surface the retarded electron–electron interaction is not known in closed form and hence one has to resort to certain limiting cases for which expression are available. Such a case is the long wavelength limit for a homogeneous electron gas where the Coulomb interaction is exponentially screened by an amount that depends on the density of states (per energy) $N(E_F)$ at the Fermi energy E_F , specifically the screening length is $\lambda_{TF} = 1/\sqrt{4\pi N(E_F)}$ [4,5]. Adopting this form of the electron–electron interaction we developed [6–8] a computational scheme based on the layer Korringa–Kohn–Rostoker (LKRR) method that treats the single-particle states on the level of local density approximation within the density-functional theory. The purpose of this work is to apply this method to the case of Cu(1 1 1) surface. This situation is particularly interesting as Cu(1 1 1) supports a surface state at roughly 0.5 eV below E_F , meaning that by tuning the energy

one can study the case of a two-dimensional (the surface state) and three-dimensional (the d-band) system.

To disentangle the emission from the surface state from that from the bulk states, the photon energy should not exceed twice the surface state energy plus the kinetic energy of the two electrons. Without this restriction, as soon as the photon energy is large enough to excite the bulk states, both bulk and surface states are contributing to the DPE.

2. Outline of the model

Within our model the single-particle properties are captured on the level of up-to-date calculations of single-photoemission (SPE) (see ref. [9]), meaning that surface and bulk states as well as photoelectron diffraction are incorporated. Upon the absorption of a photon with the energy ω two electrons are excited from the initial state $|1, 2\rangle$ to the final two-electron state $|3, 4\rangle$ which represents two electrons leaving the surface and eventually being detected. The initial two-electron state is written as $|1, 2\rangle = |1\rangle \otimes |2\rangle$, with the one-electron states $|1\rangle$ and $|2\rangle$ being the solutions of the semi-infinite crystal Hamiltonian. The latter consists of the sum of periodically arranged ion-core potentials $W_j(r_i)$, where j enumerates lattice sites and $i = 1, 2$ enumerates electrons. Due to the periodicity of the problem, each one-electron wave function is classifiable by eigenenergy, the spin value and the parallel component of the wave vector: $|i\rangle = |E_i, \mathbf{k}_i^{\parallel}; \sigma_i\rangle$, $i = 1, 2$. The final state is written as $|3, 4\rangle = |\tilde{3}\rangle \otimes |\tilde{4}\rangle$ where the tilde indicates the addition

* Corresponding author.

E-mail address: jber@mpi-halle.de (J. Berakdar).

of the wave-vector dependent term $U(r_i, \lambda; \mathbf{k}_1, \mathbf{k}_2)$ to the lattice potential. For further details we refer to ref. [7]; here we underline that the total potential is parametrically dependent on the final-state wave vectors of the outgoing electrons as to mimic (quantitatively) the directional behavior of the electron–electron interaction. In particular, when two electrons are emitted with \mathbf{k}_1 being close to \mathbf{k}_2 the DPE signal is suppressed. In the opposite case, when electrons have very different wave vectors the term U becomes negligible and the final-state total potential remains the same as that for the initial state. This immediately reduces the DPE matrix element to the separable case: $\langle 3, 4 | \Delta | 1, 2 \rangle = \langle 3 | \Delta | 1 \rangle \cdot \langle 4 | 2 \rangle$ (Δ stands for the two-electron dipole operator), giving zero value because of the last integral involving orthonormal states. Between these two limits of very close and very different asymptotic final-state wave vectors \mathbf{k}_1 and \mathbf{k}_2 , the DPE events are realized.

3. Angular patterns

It is instructive to inspect at first the single-particle features of the Cu(1 1 1) surface. Fig. 1 shows the layer-resolved density of electronic states. The surface state located ~ 0.5 eV below E_F (at $k_{\parallel} = 0$) diminishes quickly away from the surface and it constitutes an example for a two-dimensional electron gas. With the increase of k_{\parallel} , the surface state position moves towards higher energies, and at some tilt angle it crosses the Fermi level. The photoemission from the Fermi level indeed shows the in-plane circle $k_x^2 + k_y^2 = \text{constant}$, which has been observed in angle-resolved photoemission experiments (see ref. [10]), as well as in our calculations (see Fig. 2). Here we use the same photon energy ($\omega_{\text{HeI}} = 21.2$ eV) as in the experiment. The electrons reside initially at the Fermi level. Fig. 2 represents the so-called Fermi-surface map at the Cu(1 1 1) surface state. It has an almost circular shape, however, weak reflexes of the three-fold symmetry of the (1 1 1)-face are visible.

Now we ask what happens to this circle if the second electron is involved. Consider DPE in the geometry where one electron is emitted in the entire hemisphere and another one is fixed in

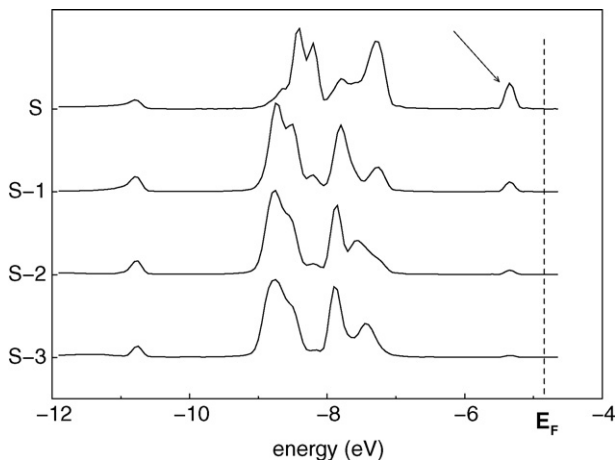


Fig. 1. Cu(1 1 1) layer-resolved density of states at $k_{\parallel} = 0$. Arrow marks the position of the surface state, which decays with the layer depth. The label S marks the number of the layer enumerated from the outmost layer $S = 0$.

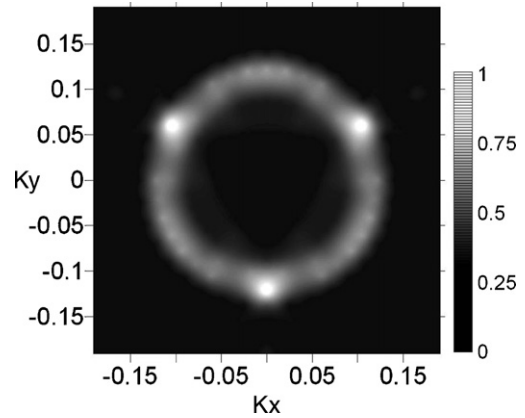


Fig. 2. Angle-resolved SPE intensity $I(k_x, k_y)_{E=E_F}$ from the Cu(1 1 1) surface state at $\hbar\omega = 21.2$ eV. The light is unpolarized.

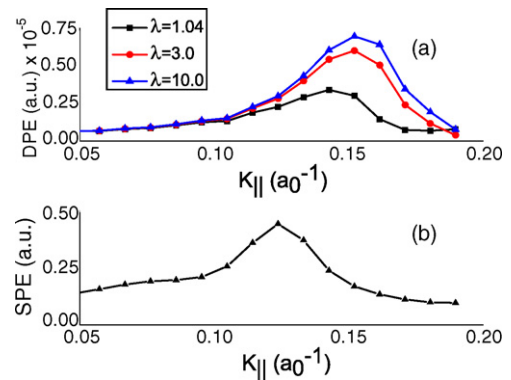


Fig. 3. (a) DPE intensity for $\hbar\omega = 42.4$ eV, $E_1 = E_2 = 16.25$ eV. One of the electrons is emitted along the surface normal direction ($(\theta, \phi) = (0, 0)$). The second electron spans the hemisphere above the surface. The resulting angular distribution is averaged over the angle ϕ and is plotted as a function of θ . (b) The SPE intensity from Fig. 2, averaged over the azimuthal angle. A variation of θ corresponds to a variation in k_{\parallel} .

the direction normal to the surface. Both electrons have the same kinetic energy which is set equal to the photoelectron energy in Fig. 2. The resulting almost isotropic distribution is averaged over the azimuthal angle and the DPE intensity is plotted as a function of the electron polar emission angle (with respect to the surface normal), which is equivalent to changing the electron parallel momentum Fig. 3. Three different values of the screening length are considered: $\lambda = 1.04a_0$ is the Thomas–Fermi value [4], another two describe less effective screening (e.g. for $\lambda = 3a_0$ two electrons are allowed to interact appreciably at a distance of the order of the lattice constant). Qualitatively, the presence of the repulsive center (fixed electron) will expand the original circle. This effect depends on λ (the less the interaction is screened, the more noticeable is the shift of the circle position towards larger angle). A corresponding measurement yields thus an estimate of the value of the screening length.

4. Energy-sharing mode

The energy correlation within the electron pair emitted in a DPE process is conventionally studied by means of the energy-

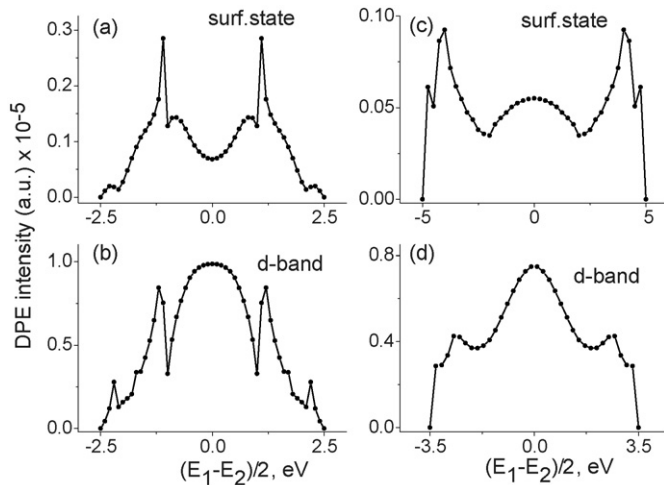


Fig. 4. DPE energy sharing distributions from the surface state and the d-band of Cu(1 1 1) for $\lambda = 1.04a_0$: (a) $\hbar\omega = 15.7$ eV, $E_{\text{tot}} = 5$ eV, $\theta_1 = -\theta_2 = 20^\circ$; (b) $\hbar\omega = 19$ eV, $E_{\text{tot}} = 5$ eV, $\theta_1 = -\theta_2 = 20^\circ$; (c) $\hbar\omega = 21$ eV, $E_{\text{tot}} = 10$ eV, $\theta_1 = -\theta_2 = 30^\circ$; (d) $\hbar\omega = 21$ eV, $E_{\text{tot}} = 7$ eV, $\theta_1 = -\theta_2 = 30^\circ$.

sharing distributions (ESD) (cf. Fig. 4). The emission directions of the two electrons are fixed and the final-state energies are chosen such that their sum $E_{\text{tot}} = E_1 + E_2$ is always constant. The ESD provides information on which combinations of kinetic energies lead to a strong DPE signal. At the edges, where one of the electrons has zero energy, the DPE cross-section drops to zero due to kinematical reasons [6]. In general, the shape is determined by the density of single-particle electronic states and by electronic correlation effects as well as by interference effects of the waves that are coherently scattered from the ionic sites. In general, it is not possible to disentangle these effects.

By appropriately choosing the photon energy, it is possible to create electron pairs originating (i) only from the surface state or (ii) both from the surface state and the d-band. Fig. 4 contrasts these two cases: electrons are emitted symmetrically (a and b) at $\pm 20^\circ$ to the surface normal; (c and d) at $\pm 30^\circ$ to the surface normal, the photon is polarized parallel to the surface in the plane of the detectors. Comparing the emission from the surface state and from the bulk in Fig. 4 a and b, respectively, one notes a strong difference in the region comparable kinetic energies. The surface state results in a lower emission cross-section at equal electron energies, in contrast to the emission characteristic from the bulk state. The total electron pair energy $E_{\text{tot}} = 5$ eV is constant for both cases, which suggests that final-state effects (which depends in the first place on the vacuum electron energies) are not very important, and the difference may arise from the combination of the following aspects: (i) strong localization of the

z-component of the surface-state wave function, (ii) different screening properties in two and three dimensions and (iii) particular features in the momentum-resolved densities of the surface state and the d-band. Similar trend is present in another pair of ESDs, see Fig. 4 c and d. Here it is the photon energy which is constant ($\omega = 21$ eV), therefore $E_{\text{tot}} = 10$ eV and $E_{\text{tot}} = 7$ eV for (c) and (d), respectively. In general, the DPE intensity is always higher for the bulk emission because of the higher density of states. It is also higher for smaller tilt angle (20° versus 30°), if we compare (a) to (c), and (b) to (d). In general we can say that the ESD from the surface states follows a behavior similar to that expected from the independent electron model [3] while for DPE from the d-band the predictions of the free-electron model [3] are inappropriate.

5. Conclusions

We have applied the DPE model calculations to the Cu(1 1 1) surface, for which the surface state and the d-band energy regions provide two different systems, as far as the dimensionality and the strength of the electron–electron interaction are concerned. We showed that the angular isotropy of the emission from the surface state, related to its almost free-electron behavior along the surface, manifests itself also in the DPE angular distributions. Since the latter are shown to be sensitive to the value of λ , the angle-resolved DPE measurements give access to information on the screening properties. As for the behavior of the energy correlation, prominent differences are found between the bulk and the surface-state DPE, mainly in the region where the photoelectrons have comparable kinetic energies.

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