ELECTRON-PAIR EMISSION FROM SOLIDS AND CLEAN SURFACES UPON ELECTRON AND PHOTON IMPACT

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INTRODUCTION

An electronic system externally distorted by photons or particle impact can integrally respond by the simultaneous emission of two electrons into the vacuum. The spectra of these correlated electron pairs carry various information depending on the scattering geometry and the initial conditions from which the pairs originated.

E.g., if the final-state electrons are generated following an isolated, direct binary collision between the fast incoming electron and one of the electrons of the target then the spectra can be related to the one-particle spectral momentum density of the initially bound electronic state [1, 2, 3, 4]. Correspondingly, the calculations done in this case deal essentially with the electronic structure of the target rather than with the collision process as such.

In contrast, addressing the collisional and dynamical aspects of the reaction becomes inevitable at lower energies [5, 6]. A particularly challenging prototype of this low-energy scattering is the emission of correlated electron pairs from semi-infinite solids upon electron impact. Such experiments have recently been performed [7, 8, 9] in the back-reflection mode in which the electron gun and the vacuum-electron detectors are in the same half space (cf. Fig.1a).

As shown in this work, the dominant ejection process of the electron pair in the back-reflection mode can be viewed as follows: We regard the electron pair as a "quasi-particle" (an uncondensed Cooper pair). The excitation process of the pair into the vacuum occurs then via diffraction of the quasi particle from the crystal potential. Diffraction conditions, equivalent to the von-Laue conditions for the Bragg scattering, can be derived. As presented here, those conditions are substantiated by experimental findings. The intensity of the diffracted spots are dictated by the scattering dynamic, in particular by the inter-electronic interaction.

The second topic treated here is the simultaneous emission of an electron pair upon single photon absorption. This reaction is markedly different from the case in which the pair is ejected following electron impact. In the photon case the two electrons are bound in the initial state. More importantly, an emission of the photo-electron pair implies a correlated motion of the pair in the initial and/or final channel [10].

In the treatment of angular-resolved ultraviolet single-photoemission spectroscopy (ARUPS) many aspects of the single photoelectron emission process are described within single-particle

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picture, as a transition from an occupied one-electron orbital to a state describing the propagation of the photoelectron. The complicated many-body nature of the solid is then collectively subsumed in the screening and decay of the photoelectron and the hole left behind. These screened, decaying quasiparticles can still be described by a single-particle wave equation. Experimental evidence for many-body effects shows up as subsidiary features in the photoelectron spectra [11, 12].

In contrast, a simultaneous two-orbital excitation by one photon is prohibited if *spatial* coupling between these orbitals is absent. Therefore dealing with the photo electron pair emission, the description of the electron-electron interaction must go beyond regarding it as a collective, spatially independent perturbation of the single-particle orbitals. Thus it seems worthwhile to employ the photo double emission (PDE) as an investigative tool for strongly correlated systems, such as Mott insulators, ferromagnetic materials with d- and f-levels, and high temperature superconductors. In fact as shown in this paper, to some extent, the PDE can be regarded as a single photoemission of a "quasi particle" formed by the electron pair. In addition to the known features of ARUPS, the spectra of this photo-emitted "quasi particle" reveal a dependence on the pair's internal degree of freedom that characterizes the mutual interaction of the two emitted electrons. Therefore, PDE experiments, which have already been conducted [13], are expected to provide a new and direct insight into the influence of electronic correlation on the initial and final many-body states.

ELECTRON-PAIR EMISSION UPON ELECTRON IMPACT

The probability for two electrons to emerge into the vacuum with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ following the impact of a projectile electron with wave vector $\mathbf{k}_0$ is related to the transition amplitude $\mathcal{T}$. This amplitude is, to leading order, a sum of direct electron-electron scattering T_{ee} and an amplitude involving the scattering from the crystal potential T_{ec} [6], i. e.

$$\mathcal{T} = T_{ee} + T_{ec}. \tag{1}$$

To elucidate the quasi-particle nature of the pair emission it is instructive to canonically transform to a momentum space spanned by $\mathbf{K}^- \otimes \mathbf{K}^+$ where $\mathbf{K}^+ = \mathbf{k}_1 + \mathbf{k}_2$ is the center-of-mass momentum of the pair, and $\mathbf{K}^- = (\mathbf{k}_1 - \mathbf{k}_2)/2$ characterizes the internal degree of freedom of the pair. The electron-electron direct scattering amplitude has the form

$$T_{ee} = \langle \mathbf{K}^-, \mathbf{K}^+ | W_{ee} | \mathbf{k}_0, \chi_{\epsilon(\mathbf{k})} \rangle. \tag{2}$$

where $|\mathbf{k}_0\rangle$ is the state vector of the incident electron. The bound state with energy ϵ and wave vector $\mathbf{k}$ is represented by $|\chi_{\epsilon(\mathbf{k})}\rangle$ and W_{ee} is a screened Coulomb scattering potential. The transition amplitude T_{ec} that describes scattering from the semi-infinite crystal can be deduced to

$$T_{ec} = \iint d^3\mathbf{p}\, d^3\mathbf{q} \langle \mathbf{K}^-, \mathbf{K}^+ | W_{ee} g_{ee}^- | \mathbf{p}, \mathbf{q} \rangle \langle \mathbf{p} | W_{ec} | \mathbf{k}_0 \rangle \langle \mathbf{q} | \chi_{\epsilon(\mathbf{k})} \rangle. \tag{3}$$

Here g_{ee}^- is the propagator in the electron-electron Coulomb potential, W_{ec} is the interaction potential between the projectile and the lattice and $|\mathbf{q}\rangle \otimes |\mathbf{p}\rangle$ is a complete set of plane waves. Assuming non-overlapping muffin-tin ionic potentials V^{ion} ($W_{ec} = \sum_i V_i^{ion}$), the form factor $\tilde{W}_{ec} := \langle \mathbf{p} | W_{ec} | \mathbf{k}_0 \rangle$ is reduced to

$$\tilde{W}_{ec} = \frac{N\sqrt{2\pi} f}{A_{uc}} \sum_{\ell} e^{-iK_z r_{\perp,\ell}} \sum_{\mathbf{g}_\parallel} \delta^{(2)}(\mathbf{g}_\parallel - \mathbf{K}_\parallel) \tilde{V}^{ion}(\mathbf{K}). \tag{4}$$

In Eq. (4) $\tilde{V}^{ion}(\mathbf{K})$ is the Fourier transform of V^{ion}, N is the number of ionic cores illuminated by the electron beam, A_{uc} is the two-dimensional unit cell, $\mathbf{g}_\parallel$ is the surface reciprocal

lattice vector, ℓ enumerates the atomic layers with shortest distance $r_{\perp,\ell}$ with respect to the origin, $\mathbf{K} = \mathbf{p} - \mathbf{k}_0$, and $f = \exp(i\mathbf{p}\cdot\mathbf{r}_1)$ with $\mathbf{r}_1$ refering to the position of the bound electron.

Using Bloch's theorem the amplitudes T_{ec} and T_{ee} can be formulated as

$$T_{ec} = C\sum_{\ell,\mathbf{g}_{||}} \delta^{(2)}[\mathbf{g}_{||} - (\mathbf{K}^+_{||} - \mathbf{K}_{0,||})]\,\mathcal{L}(\mathbf{g}_{||},\ell,\mathbf{K}^+,\mathbf{K}^-,\mathbf{k}) \tag{5}$$

$$T_{ee} = \delta^{(2)}(\mathbf{K}_{0,||} - \mathbf{K}^+_{||})\,\mathcal{L}'. \tag{6}$$

In Eqs. (5,6) $\mathbf{K}_0 = \mathbf{k}_0 + \mathbf{k}$ is the initial wave vector of the pair. The functions $C, \mathcal{L}, \mathcal{L}'$ depend on the description of the momentum-space wave function $\langle\mathbf{q}|\chi_{\epsilon(\mathbf{k})}\rangle$ of the bound electron and for a jellium-state momentum distribution can be evaluated in closed form [6]. The structure of Eq. (5) has the following implications:

1) The Bragg-like diffraction condition, expressed by the delta function, involves the pair center-of-mass wave vector only. This can be interpreted as a diffraction of a quasi particle located at the pair's center of mass when the parallel component of its wave vector is changed by $\mathbf{g}_{||}$ during the collision. Note that for Bragg scattering, diffraction occurs when the momentum change of the incident electron equals to a multiple of $\mathbf{g}_{||}$. The decisive difference to the pair's diffraction is that a fixed $\mathbf{K}^+$ does not imply a fixed $\mathbf{k}_1$ and/or $\mathbf{k}_2$ since a momentum exchange of the two electrons (the internal coordinate $\mathbf{K}^-$ changes then) does not necessarily modify $\mathbf{K}^+$. Therefore, a definite change in $\mathbf{K}^+$ does not fix the amount of momentum change of the incoming projectile.

2) The positions of the diffraction spots is dictated by $\mathbf{K}^+$. The intensity of these spots is determined by the function $\mathcal{L}$ that depends on $\mathbf{K}^-$, i. e. $\mathcal{L}$ in influenced by the strength of electronic correlations (in momentum space W_{ee} depends only on $|\mathbf{K}^-|$).

3) The diffraction spots are blurred by the distribution of the wave vector $\mathbf{k}_{||}$ of the initially bound Bloch electron, even in the case where $\mathbf{K}^+$ and $\mathbf{k}_0$ are sharply resolved.

To substantiate the above statements we compare the theoretical results with experiments performed on Cu(001) mono-crystal. The cross section is then proportional to $|\mathcal{T}|^2$ (Eq. 1). An integration over $\mathbf{k}_{||}$ (weighted with the density of states) is, however, necessary as it is not experimentally resolved [6]. In the experiment , for a given incident energy E_i and total excess energies $E_{tot} = (k_1^2 + k_2^2)/2$ of the pair, the electrons' energy sharing is scanned. As shown in Fig. 1a, $\mathbf{k}_0, \mathbf{k}_1, \mathbf{k}_2$ lie in the $x-z$ plane, i. e. $\mathbf{K}^+_{||}$ possesses only one non-vanishing component K^+_x along the x axis. As indicated above, it is this component that is relevant for the pair diffraction and hence we investigate the energy sharing as function of K^+_x. Since $E_{tot} = {K^+}^2/(2m) + {K^-}^2/(2m_\mu)$, where $m = 2$ is the total mass of the pair and $m_\mu = 0.5$ is their reduced mass, the value of K^+_x is, in the case of Fig.1a, restricted to

$$-\sin\alpha\sqrt{2E_{tot}} \le K^+_x \le \sin\alpha\sqrt{2E_{tot}}. \tag{7}$$

Note that the normal of the surface is a cylindrical symmetry axis of the scattering plane while the crystal structure is invariant under 180^o rotation. Therefore, the spectrum, depicted in Fig.1b, is symmetric with respect to $K^+_x = 0$.

The positions of the first diffracted beams are denoted by $(-1,0)$ $(1,0)$ in Fig.1b (assuming stationary bound electron ($\mathbf{k}_{||} = 0$). The onset of the (1,0), and (-1,0) diffraction spots are clearly identified in Fig.1b. The abrupt decrease of these peaks at the wings is due to the cut-off condition (7). The structure in the middle is due to the specular beam (0,0). This is evident from the incoherent contributions $|T_{ee}|$ and $|T_{ec}|$ to the total transition amplitude (1).

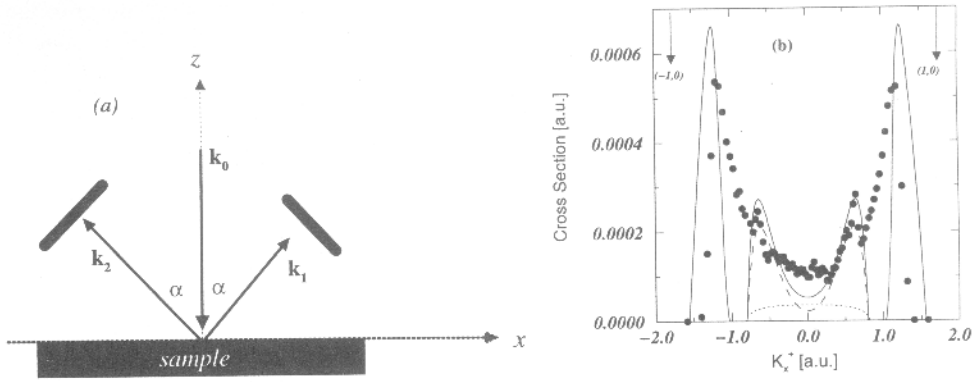

Fig.1. **(a)** schematic representation of the experimental setup; $\mathbf{k}_1, \mathbf{k}_2$ denote the momenta of the escaping electrons under an angle α with respect to $\hat{\mathbf{z}}$ and $\mathbf{k}_0$ is the incident electron momentum. **(b)** For a fixed incident energy $E_i = 85\,eV$ and fixed excess energy $E_{tot} = 79\,eV$, the excess energy-sharing of the escaping electrons is depicted as function of K_x^+. The experiment has been performed on a Cu(001) mono crystal at a fixed $\alpha = \cos^{-1}(\hat{\mathbf{z}} \cdot \hat{\mathbf{k}}_1) = \cos^{-1}(\hat{\mathbf{z}} \cdot \hat{\mathbf{k}}_2) = 40^o$. The cross sections calculated with the amplitude $|T_{ee}|$ (Eq. 6) only (dotted curve) and with $|T_{ec}|$ only (dashed) curve are shown along with that predicted by the coherent sum $|T|$ (Eq. 1). The calculations are performed for infinite energy and angular resolution of the detectors. The arrows indicate the positions of the (1,0) and (-1,0) diffracted beams. The theoretical (-1,0) and (1,0) diffracted peaks are scaled down by 2. The experiment are due to Ref.[14].

The major contribution originates from T_{ec} (Eq. 5), i. e. from scattering events involving the pair's back-reflection from the crystal.

Remarkably, the finite-size of the amplitude T_{ee} (Eq. 6) still results in an interference of this amplitude with T_{ec}. The contribution of $|T_{ee}|$ (Eq. 6) to the (-1,0) and (1,0) peaks vanishes identically, as evident from Eq. (6). This underlines the identification of these peaks as (-1,0) and (1,0) diffraction beams of the pair.

Inspecting the sum over ℓ in Eq. (5) we deduce that in the case of Figs. 1(b) correlated pairs are predominantly generated in the two top-most atomic layers.

In view of applying this technique to electronic-structure investigations, as suggested by Refs. [4], "clean" diffracted beams are needed. As illustrated in Fig. 1(b), the (-1,0) and (1,0) diffracted peaks seem most suitable for this purpose as contamination from other non-diffractive processes are excluded.

ONE-PHOTON ELECTRON-PAIR EMISSION

The cross section for the pair's emission upon photon impact is derived to [10]

$$\frac{d\sigma}{d^3\mathbf{k}_2\, d^3\mathbf{k}_1} = 4\pi^2 \frac{\alpha_c}{\omega} \sum_{M_f} \frac{1}{2J_i+1} \sum_{M_i} \int\int d^3\mathbf{k}'_1\, d^3\mathbf{k}'_2\, \rho(\mathbf{k}'_1) F(\mathbf{k}'_1, T) \rho(\mathbf{k}'_2) F(\mathbf{k}'_2, T)\, |M_{fi}|^2 \delta(E_i - E_f) \tag{8}$$

α_c is the fine-structure constant and ω is the photon frequency. Eq. (8) averages over the initial magnetic sublevels M_i, and sums over the magnetic sublevels M_f of the hole states with orbital momentum J_i. The one-particle density of states at the temperature T is referred to as $\rho(\mathbf{k}'_j)$, $j = 1, 2$ and $F(\mathbf{k}'_j, T)$ is the Fermi distribution and $\mathbf{k}'_j$, $j = 1, 2$ are Bloch wave

vectors of the initially bound electrons. E_f is the final-state energy. The initial total energy E_i can be estimated assuming the (valence) band of the pair as being formed of independent bands of the single electrons. For DPE from core state, it can be shown that the optical transition amplitude with $k_j \gg k'_j$, $j = 1, 2$ can be written as [10]

$$|M_{fi}|^2 = |C|^2 \delta^{(3)}_{(\mathbf{K}_0^+ - \mathbf{K}^+, \mathbf{g})} |\hat{\mathbf{e}} \cdot \mathbf{K}^+|^2 |\tilde{\phi}_i(\mathbf{K}^+, \mathbf{K}^-)|^2 \tag{9}$$

where $\mathbf{K}_0^+ = \mathbf{k}'_1 + \mathbf{k}'_2$, $\mathbf{g}$ is a bulk reciprocal lattice vector, $\hat{\mathbf{e}}$ is the photon's polarization vector, C is a flux factor, and $\tilde{\phi}_i$ is the double Fourier transform of the electronically *correlated* initial state. The photo electron-pair diffraction admits the interpretation of a quasi-particle diffraction as demonstrated above for the electron-impact case. The optical selection rules are expressed in the scalar product $\hat{\mathbf{e}} \cdot \mathbf{K}^+$ that suggests an absorption of the photon by the center of mass of the pair.

For DPE from delocalized electronic valence states the situation is more complex. A treatment of this case can be found in Ref. [10] where numerical example are presented.

To the cross section (8) the contribution of an additional channel should be added. In this channel the pair is generated via a single photoabsorption followed by an (e,2e) process.

Future studies on this subject include the PDE from high temperature superconductors and magnetic thin films.

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