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# Electron pair emission from a W(001) surface: photon versus electron excitation

M Muñoz-Navia, C Winkler, R Patel, M Birke, F O Schumann and J Kirschner

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

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## Abstract

The electron pair emission from a W(001) surface was studied using a coincidence time-of-flight spectrometer. The aim of this study was to compare the pair emission upon electron impact and upon photon absorption. The energy distributions are markedly different for these two experiments. From this we conclude that the photon-stimulated pair emission carries a significant contribution from a double photoemission process, while the process of first creating a photoelectron, which in a subsequent collision leads to pair emission, is of less importance.

## 1. Introduction

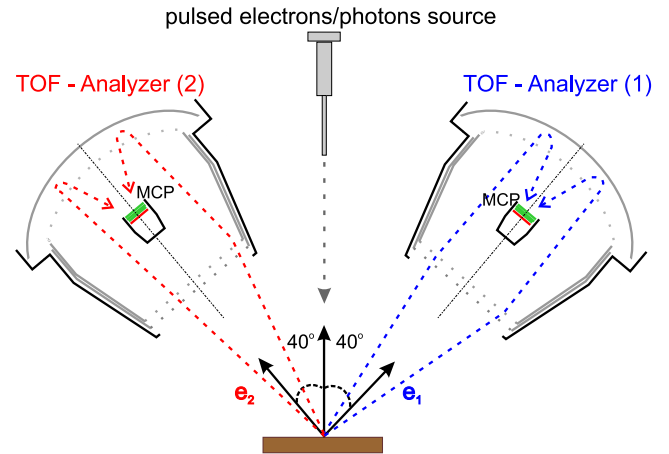
The description of electronic properties of solids via an independent particle picture has proven to be an adequate framework. Admittedly, the simplest model, namely the free electron gas, captures essential properties of the many-body system like the temperature dependence of the electronic specific heat. However, the independent particle picture can only be an approximation, since the electrons actually interact. On the microscopic scale the Pauli principle and the Coulomb interaction are responsible for the mutual influence. This electron–electron interaction is responsible, for example, for many-body effects like magnetism and superconductivity [1–3]. An experimental tool which allows us to access the electronic properties of materials is photoemission. Recent improvements on the experimental side have made it possible to observe aspects of the electron–electron interaction, which show up as so-called kinks in the energy versus momentum distributions (or dispersion  $E(k)$ ). However, the discussion of these results are usually performed within an effective single-electron picture. A natural next step, which goes beyond an effective single-particle description, is to probe the electron pair emission from surfaces. If we excite the surface with a primary electron, we term this experiment ( $e$ ,  $2e$ ). If we select a photon for excitation we label it a ( $\gamma$ ,  $2e$ ) experiment. In our previous work we have demonstrated that in both cases experimental access to an important concept of modern solid state theory, namely the exchange–correlation hole, is possible [4–10]. When one considers the ( $\gamma$ ,  $2e$ ) process it becomes apparent that two possible pathways for pair emission exist [4, 11, 12]. On the one hand, a single photon can be absorbed, which is accompanied by the emission

of an electron pair. This is regarded to be a direct double photoemission process (DPE). This process is only possible if the two electrons are correlated [12]. A simple picture illustrating the plausibility of this point is to consider two valence electrons, which initially move independently of each other. Once they collide, their motion through the crystal is correlated. Within the dipole approximation the photon is absorbed by one electron, but by virtue of the electron correlation the second electron is affected as well, which can lead to the emission of a pair. Without correlation between the electrons only single photoemission is possible. This leads us immediately to the other pathway. It is conceivable that the photon is absorbed by a single electron, resulting in the creation of a photoelectron. After a series of collisions, in which it loses its initial phase, it may collide with another electron, which creates an electron pair. The process may be called an internal ( $e$ ,  $2e$ ) event. The timescale over which these processes take place is of the order of 10 fs. This time needs to be compared with the time resolution of our instrumentation, which is currently of the order of 1 ns. Therefore it is experimentally not possible to separate these different pathways directly. Recent development in laser technology have shown that attosecond light pulses can be generated [13], which may allow us in the future to separate the two pathways directly. It is expected that both scenarios make comparable contributions to the total ( $\gamma$ ,  $2e$ ) intensity, because in both cases the underlying interactions, namely electron–electron and electron–photon interactions, appear in the same order [14]. In coincidence experiments one has to operate at low primary fluxes in order to reduce the contributions of so-called random coincidences. The random coincidences vary quadratically with the primary flux, while the true coincidences

vary linearly with the flux. An operational compromise is to choose the primary intensity such that true and random coincidences make about the same contribution to the total coincidence signal. In principle it is possible to determine the absolute cross section for pair emission. This requires us to operate at even lower primary fluxes. It would be appealing to measure these cross sections for the two different excitations. However, a direct comparison between  $(e, 2e)$  and  $(\gamma, 2e)$  signals is not easily possible. It has to be remembered that the primary photon will travel much deeper into the sample compared to the primary electron. This means essentially that, although the photon can generate a pair, it is too far away from the surface to be emitted. Because of this, a separation of the DPE channel versus the internal  $(e, 2e)$  channel on grounds of the intensity levels is not possible. It is important to point out that both experiments have comparable probing depths, because two low kinetic energy electrons have to be detected in coincidence. As an attempt to unravel the different contributions to the  $(\gamma, 2e)$  intensity, we performed  $(e, 2e)$  and  $(\gamma, 2e)$  experiments on the same sample under otherwise identical experimental conditions. In a simplified picture one would expect that similar energy distributions for both experiments are indicative of a dominant contribution of internal  $(e, 2e)$  rather than a genuine DPE contribution. We studied a W(001) surface and find that the energy distributions of the two type of experiments display distinctive differences.

## 2. Experimental details

Electron coincidence experiments were performed utilizing a pair of reflection time-of-flight (TOF) detectors equipped with multichannel plates (MCP). A schematic view of the experimental set-up is displayed in figure 1. Electrons entering the detector are reflected by an electrical mirror and projected into the active area of the channel plates. This concept has been indicated by example trajectories, which have been included in figure 1. A more detailed description including the performance of this instrument can be found elsewhere [15]. The sample is mounted such that the primary electrons or photons are incident along the surface normal. The electron optical axes of both detectors include an angle of  $80^\circ$ , while each of them makes an angle of  $40^\circ$  with respect to the surface normal. The angular acceptance of each detector is  $25^\circ$ , which results in a total solid angle of detection of  $\Omega \approx 1$  sr. In the  $(\gamma, 2e)$  experiments, we used as a pulsed light source the synchrotron storage ring BESSY II operated in single-bunch mode. In this mode a light pulse hits the sample every 800 ns. Our experiment was connected to the beamline CP-NIM, which delivers circularly polarized light in the energy range 5–40 eV [16]. The BESSY bunch marker was taken as a time reference for the time-of-flight measurements. The  $(e, 2e)$  experiments were performed with a home-built electron gun and the beam could be switched on or off via applying voltages to deflection plates. The voltages were supplied by a pulse generator. From the time-of-flight we can compute the kinetic energy due to the known flight path. All quoted kinetic energies are with respect to the vacuum level. In the following we label the kinetic energies of the

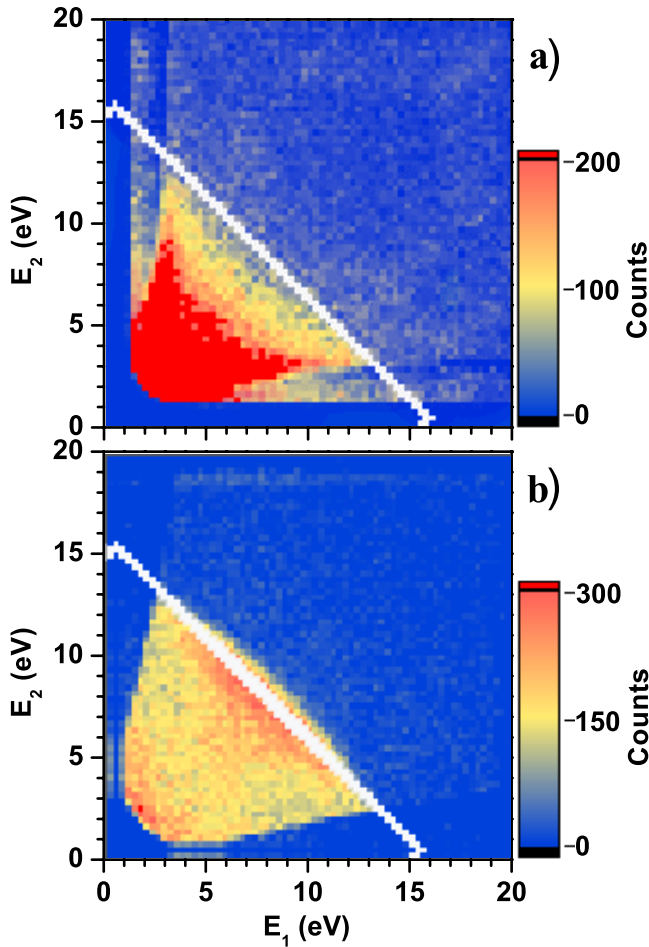


**Figure 1.** Schematic view of the experiment. The primary electron or photon beam hits the sample along the surface normal, while the electron optical axis of the two detectors has an angle of  $40^\circ$  with respect to the surface normal. The detectors' optical axis and the primary beam are in the drawing plane. For each detector we show example trajectories to explain the idea of the electron mirror.

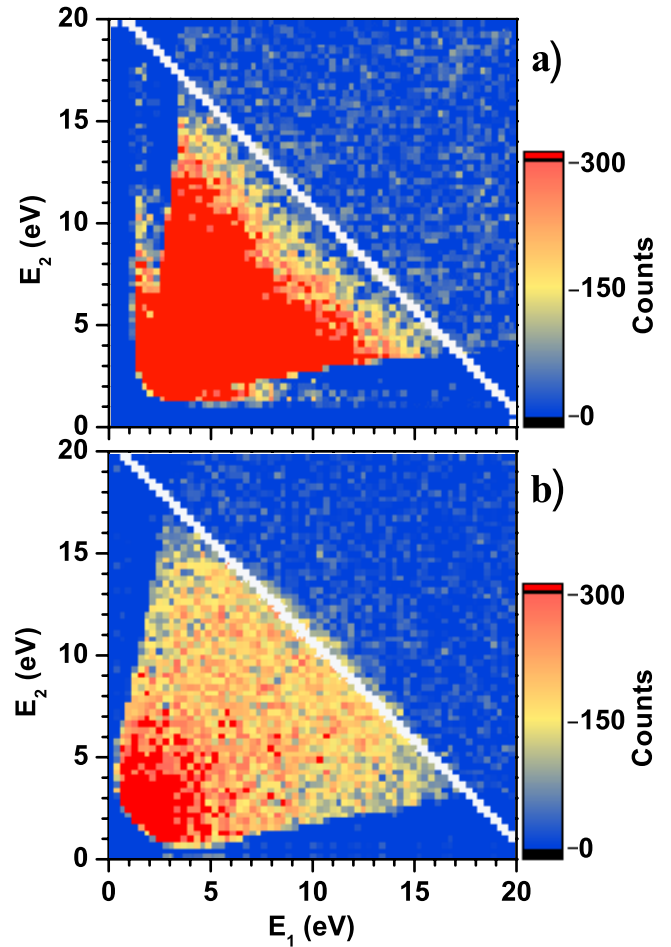
electrons constituting the pair as  $E_1$  and  $E_2$ , respectively. The time resolution we could achieve resulted in an energy resolution of  $\Delta E \approx 0.5$  eV for an electron with a kinetic energy of 20 eV. With the help of a coincidence circuit it was ensured that only electron pairs were registered. The above-described electron spectrometers were part of a ultra-high vacuum system equipped with standard surface science instrumentation. The experiments were performed at a base pressure in the  $10^{-11}$  mbar range.

## 3. Results and discussion

The W(001) surface was cleaned by standard procedures, which involved first heating the crystal to a temperature of  $\approx 2000^\circ\text{C}$  for approximately 1 h in a  $5 \times 10^{-8}$  mbar oxygen atmosphere to remove carbon and CO. Subsequently, the crystal was flashed several times up to  $\approx 1000^\circ\text{C}$  to remove oxygen. The treatment was repeated initially until the surface was judged to be clean by Auger electron spectroscopy and no traces of carbon migrating from the bulk were found. The sample was periodically (about every 45 min) heated up to  $\approx 1000^\circ\text{C}$  between measurements to remove adsorbed gases from the sample surface. The coincidence count rate was of the order of 1 cps, while the individual detectors showed a rate of about 1000 cps. For the  $(\gamma, 2e)$  experiment a total of four weeks of beamtime was available, which amounts to the maximum single-bunch beamtime available over the duration of a year at BESSY II. The  $(\gamma, 2e)$  spectra were obtained by measuring over identical time periods with either left or right circular polarized light. The individual spectra showed no difference. Therefore we combined these two datasets. The sum energy  $E_{\text{sum}}$  of a pair is defined to be  $E_1 + E_2$ , where  $E_1$  and  $E_2$  are the kinetic energy of the individual electrons. In order to facilitate comparison between  $(e, 2e)$  and  $(\gamma, 2e)$  experiments it is useful to ensure similar kinematics. More precisely, it is advantageous to keep the maximum sum energy



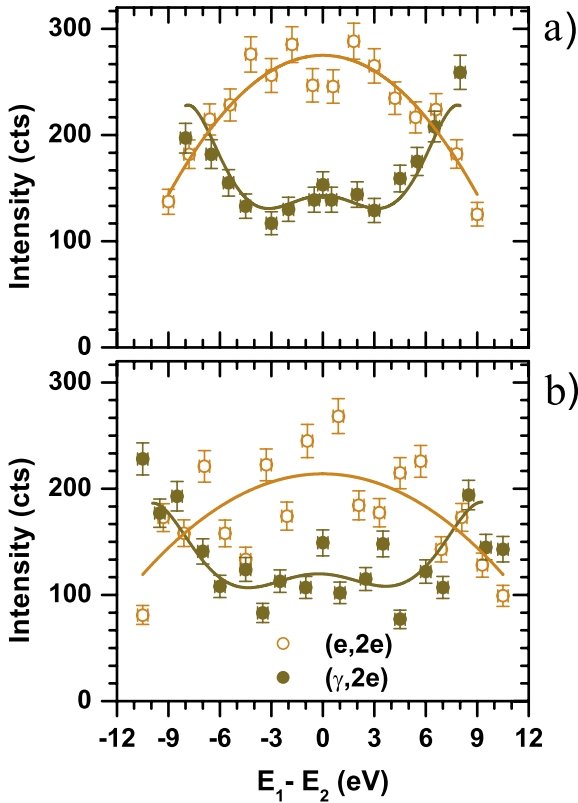
**Figure 2.** In both panels we plot the 2D energy distributions of the coincidence intensity. Panel (a) shows the experimental data for a ( $\gamma$ , 2e) experiment with  $h\nu = 25$  eV, while the results for an (e, 2e) experiment with  $E_p = 20$  eV are shown in panel (b). In both experiments the maximum sum energy of the electron pair is 15 eV. The white diagonal line (to guide the eye) in both panels indicates this energetic position of the events.



**Figure 3.** In both panels we plot the 2D energy distributions of the coincidence intensity. Panel (a) shows the experimental data for a ( $\gamma$ , 2e) experiment with  $h\nu = 29.5$  eV, while the results for an (e, 2e) experiment with  $E_p = 25$  eV are shown in panel (b). In both experiments the maximum sum energy of the electron pair is 20 eV. The white diagonal line in both panels indicates this energetic position of the events.

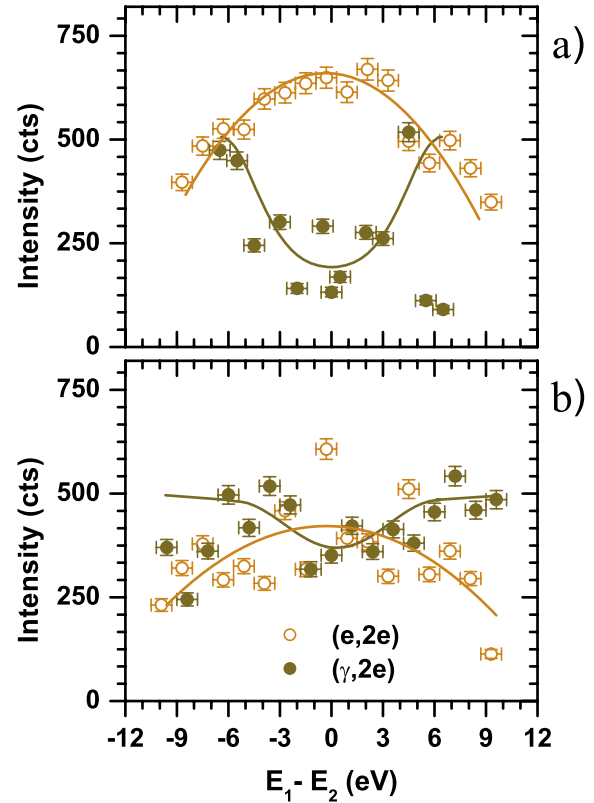
$E_{\text{sum}}^{\text{max}}$  of the electron pairs the same. Because in a ( $\gamma$ , 2e) experiment two electrons are removed from the solid we have  $E_{\text{sum}}^{\text{max}} = h\nu - 2W_A$ . In this context the photon energy is labelled by  $h\nu$  while the work function of the surface is determined by  $W_A$ . The situation is different in an (e, 2e) experiment: although two electrons leave the sample energetically the work function has to be accounted for only once, because the primary electron gains the work function when entering the sample. If  $E_p$  refers to the primary energy of the impinging electron beam, we find for  $E_{\text{sum}}^{\text{max}} = E_p - W_A$ . Using the same kinematics is ensured if we equate the two equations for the maximum sum energy. We finally obtain the relation  $h\nu = E_p + W_A$ . The work function for a W(001) surface is 4.6 eV. In the following we want to compare pair emission spectra excited with 25 eV (29.5 eV) photons and 20 eV (25 eV) primary electrons, which meet the criterion between photon energy and primary energy. In figures 2 and 3 we display the 2D energy distributions of the coincidence intensity, where the  $x$  and  $y$  axes are the kinetic energy  $E_1$  and  $E_2$ , respectively.

In order to guide the eye we have added white diagonal lines to the plots. The position of these lines mark the energetic position of the pair emission if the valence electrons originate from the highest occupied state (or the Fermi level). A first glance at figures 2 and 3 reveals that a sudden increase of the coincidence intensity occurs if one moves below this diagonal line towards smaller kinetic energies. Energy conservation demands that coincidences, which are energetically above the diagonal line, cannot come from ‘true’ coincidences, where the pair emission is triggered by the absorption of a single primary particle. The events above the diagonal line come from so-called ‘accidental’ coincidences. These arise because the number of primary particles in a pulse is determined by the Poisson statistics. Therefore it is possible that a pulse contains more than one primary particle. In this case we have to consider the possibility that each of the two particles initiates the emission of a single electron, which will also be registered by our coincidence circuit. It is clear from figures 2 and 3 that the ‘accidental’ coincidences occur with small probability.



**Figure 4.** Comparison of the sharing curves for (e, 2e) data are labelled with open circles and ( $\gamma$ , 2e) results labelled by full circles. The curves through the data points are a guide to the eye. Panel (a) includes data where  $E_{\text{sum}}$  is fixed to  $15 \pm 0.6$  eV, while  $h\nu = 25$  eV and  $E_p = 20$  eV. In panel (b)  $E_{\text{sum}}$  is fixed to  $20 \pm 0.6$  eV, while the excitation energies are  $h\nu = 29.5$  eV and  $E_p = 25$  eV, respectively.

It is immediately apparent that for (e, 2e) experiments the onset of pair emission occurs near the maximum sum energy (figures 2 and 3 panel (b)). This is also the case for primary energies of 15 eV (not shown) and also in line with previous observations [17–19]. Compared to the (e, 2e) data the ( $\gamma$ , 2e) energy distributions reveal a different result. First, the onset of pair emission does not occur near  $E_{\text{sum}}^{\text{max}}$ , but is roughly 3 eV below this. Second, for  $h\nu = 25$  eV the onset of pair emission resembles a concave curve, see figure 2(a). For equal energies of the electrons the coincidence intensity is smaller than for unequal energies. This tendency is less pronounced in figure 3(a), but can be made visible by a further analysis. For this we compute so-called sharing distributions. In this representation one selects those coincidence events for which the sum energy adopts a constant energy. This corresponds to a line, which is parallel to the white line in figures 2 and 3. These data are plotted as a function of the energy sharing which is simply the energy difference  $E_1 - E_2$ . In figure 4 we show the result for (e, 2e) experiments together with the data of ( $\gamma$ , 2e) experiments. The curves have been added to guide the eye. The symmetry of the experimental set-up suggests a symmetric curve with respect to  $E_1 - E_2 = 0$  and therefore we used polynomials with even exponents only. In figure 4(a) the values of  $h\nu$  and  $E_p$  are 25 eV and 20 eV, respectively. The value of  $E_{\text{sum}}$  was fixed to  $15 \pm 0.6$  eV. This choice ensured



**Figure 5.** Comparison of sharing curves for ( $\gamma$ , 2e) data (full circles) and (e, 2e) data (open circles). The curves through the data points are a guide to the eye. Panel (a) concerns data where photon energy and primary energy were fixed to  $h\nu = 25$  eV and  $E_p = 20$  eV, respectively. The sum energy was set to  $E_{\text{sum}} = 12.8 \pm 0.6$  eV ( $\gamma$ , 2e) and  $E_{\text{sum}} = 13.9 \pm 0.6$  eV (e, 2e). Similarly in panel (b) photon energy and primary energy were fixed to  $h\nu = 29.5$  eV and  $E_p = 25$  eV, respectively. The sum energy was set to  $E_{\text{sum}} = 17.3 \pm 0.6$  eV ( $\gamma$ , 2e) and  $E_{\text{sum}} = 18.9 \pm 0.6$  eV (e, 2e).

that essentially valence states near the Fermi level make a contribution to the coincidence intensity. The energy window we allow reflects the energy resolution of our instrument. We can clearly observe that both sharing curves are distinctively different. Whereas the sharing for the (e, 2e) experiment has a maximum for equal energies, the ( $\gamma$ , 2e) experiment displays a minimum. Panel (b) of figure 4 reflects the situation for  $h\nu = 29.5$  eV and  $E_p = 25$  eV, respectively. Setting  $E_{\text{sum}} = 20 \pm 0.6$  eV focuses on the electronic states near  $E_F$ . We note again that the ( $\gamma$ , 2e) experiment displays a minimum in the sharing curve in contrast to the (e, 2e) result. It is appealing to determine also the sharing distributions for a different value of  $E_{\text{sum}}$ . If we are interested in the contribution of valence states, which are 1.5 eV below  $E_F$ , we need to use a value of  $E_{\text{sum}} = E_{\text{sum}}^{\text{max}} - 1.5$  eV. For a proper comparison we have for the ( $\gamma$ , 2e) experiment to choose  $E_{\text{sum}} = E_{\text{sum}}^{\text{max}} - 2 \times 1.5$  eV, because two valence electrons leave the sample. The resulting sharing distributions are shown in figure 5. In panel (a) we plot the result for  $h\nu = 25$  eV and  $E_p = 20$  eV, while panel (b) shows the result for  $h\nu = 29.5$  eV and  $E_p = 25$  eV, respectively. We note that in figure 5(a) similar to figure 4 the (e, 2e) data reveal that equal energies of the electrons are preferred over unequal energies. The ( $\gamma$ , 2e) shows the

opposite in the sense that equal energies result in a lower coincidence intensity than for unequal energies. The situation becomes less clear for the data plotted in figure 5(b). There the sharing curves for (e, 2e) and ( $\gamma$ , 2e) look rather similar. Above we have compared the sharing curves for two scenarios, namely the emission from valence states near the Fermi level and from states which are  $\approx 1.5$  eV below  $E_F$ . This choice was not arbitrary. In the first case contributions from surface states are energetically possible, whereas in the second case essentially only bulk states can contribute to the coincidence intensity. This becomes more apparent if we recall some basic facts of the electronic properties of W [20–23]. The electronic density of states (DOS) of bulk W displays two regions of high density, which are located above and below the Fermi level. These correspond to bonding and antibonding d states, which display little dispersion. The region of high DOS for occupied states starts to emerge for a binding energy of 1.5 eV below  $E_F$ , while at the Fermi level itself one has a low DOS emanating from the strongly dispersing sp states. At the (001) surface these electronic states are significantly changed. It is well known experimentally and theoretically that surface states and resonances exist for this surface [20–23]. A key difference between the (e, 2e) and ( $\gamma$ , 2e) experiments is that the onset of pair emission for the latter occurs about 3 eV below  $E_{\text{sum}}^{\text{max}}$ .

Let us assume for the time being that the ( $\gamma$ , 2e) process proceeds via the ‘internal’ (e, 2e) channel. At this point we recall some well-known facts of the photoemission spectra obtained from a W(001) surface [20–22]. In the range  $h\nu = 10\text{--}40$  eV the spectra reveal prominent intensity at about 0.4 eV below  $E_F$ . This is usually ascribed to transitions which involve surface states/resonances. The photoemission intensity is strongest for  $k_{\parallel} = 0 \text{ \AA}^{-1}$ . From this discussion we expect that via photoemission a ‘primary’ beam is generated, which moves towards the surface approximately in the direction of the surface normal and has a kinetic energy of about  $h\nu$  inside the crystal. This resembles essentially the situation of the (e, 2e) experiment, where elastic scattering reverses the momentum of the primary electron such that it travels towards the surface and may encounter a valence electron leading to the emission of a pair. The photoelectron can now scatter with a valence band electron and from the (e, 2e) experiments we know that the collision with a surface state/resonance is a likely scenario, because the onset of pair emission occurs at the maximum sum energy. Consequently we expect that an ‘internal’ (e, 2e) process should lead to a high coincidence intensity at the maximum sum energy. This expectation, however, is at odds with the experimental evidence, see figures 2 and 3. For the DPE process on solid surfaces a selection rule has been derived [12, 24]. In simple terms it states that, if the sum momentum vector  $\mathbf{K}_{\text{sum}} = \mathbf{k}_1 + \mathbf{k}_2$  of the emitted electron pair is perpendicular to the polarization vector  $\mathbf{P}$  of linear polarized light, no DPE intensity can be observed. In our experimental geometry the trajectories of the electrons lie close to a plane defined by the electron optical axes of the spectrometer, i.e. the drawing plane of figure 1. Therefore the components of  $\mathbf{K}_{\text{sum}}$  stay close to this plane, too. In particular, if the kinetic energies of the individual electrons are the same and the emission angles are equal and opposite,  $\mathbf{K}_{\text{sum}}$  will be parallel to the surface

normal. If the electron energies are unequal  $\mathbf{K}_{\text{sum}}$  will have a sizeable component in the surface plane. If the incoming light hits the surface along the surface normal and the linear polarization is in the drawing plane of figure 1 we expect a reduced coincidence intensity for equal energies compared to unequal energies. This is exactly what we observe in figure 2(a) and the associated sharing curves displayed in figures 4(a) and 5(a). The fact that the polarization in our experiments is circular rather than linear does not qualitatively change our argument [24]. We conclude that (e, 2e) spectra and ( $\gamma$ , 2e) data from a W(001) surface show significant differences, which indicate that the ( $\gamma$ , 2e) process contains a sizeable contribution from a genuine DPE process.

#### 4. Summary

In this work we have compared the electron pair emission from a W(001) surface either excited by photons or a primary electron beam. The energy distributions, most notably so-called sharing curves, reveal differences. The (e, 2e) data reveal a tendency for both electrons to have the same kinetic energy, whereas ( $\gamma$ , 2e) data show a tendency to avoid equal energies. From this we conclude that a genuine DPE process makes an important contribution to the measured coincidence intensity in the ( $\gamma$ , 2e) experiments.

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