



Band-Resolved Double Photoemission Spectroscopy on Correlated Valence Electron Pairs in Metals

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Correlated valence electrons in Ag and Cu are investigated using double photoemission spectroscopy driven by a high-order harmonic light source. Electron pairs consisting of two d electrons as well as pairs with one sp and one d electron are resolved in the two-dimensional energy spectrum. Surprisingly, the intensity ratio of sp - d to d - d pairs from Ag is 3 times higher than in the self-convoluted density of states. Our results directly show the band-resolved configurations of electron pairs in solids and emphasize a band-dependent picture for electron correlation even in these paradigmatic metals.

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Since the discovery of magnetism and superconductivity, studying electron correlation has become an indispensable field of physics [1,2]. Because of the central role of the electron-electron interaction in strongly correlated materials [3] as well as its impact on molecular and single-electron devices [4,5], varieties of spectroscopies have been devoted to measure correlated electrons in solids. Among them, double photoemission (DPE) experiments analyze pairs of interacting electrons directly and have been developed progressively over several decades [6,7].

Generally, DPE on solids is challenging due to the coincidence detection of two photoelectrons. This explicitly involves a reduced joint acceptance of two spectrometers and a compromised energy resolution due to a broad, simultaneously detected energy range up to few tens eV. Moreover, to suppress accidental coincident events, the incident photon flux must be kept low [7] and the measurement time becomes long, up to several days. With a laboratory high-order harmonic light source and a pair of time-of-flight spectrometers, we recently constructed a new DPE setup [Fig. 1(a), inset] [8,9]. It allows us to reveal the band-dependent signatures of the two-particle valence spectra of Ag and Cu. Pairs of interacting valence electrons are identified according to their sum kinetic energies (E_{sum}) and specifically related to the number of participating d electrons. These two-electron E_{sum} features constitute a more intricate structure than the self-convoluted single-particle density of states and provide evidence for a distinctly band-dependent electron correlation even in these conventional metals. Because E_{sum} is a good quantum number for an electron pair instead of their individual energies, our DPE results provide valuable information regarding the electron pair configurations which go beyond the capabilities of single-particle spectroscopies.

DPE experiments were performed with s - and p -polarized light with photon energies ($h\nu$) of 32.3 and 25.1 eV,

respectively. The photoelectron pairs were analyzed by a pair of TOF spectrometers, each having a $\pm 15^\circ$ acceptance and oriented at $\pm 45^\circ$ to the sample surface normal [8,9]. Figure 1(a) shows the raw two-particle DPE histogram from Ag(001) as a function of kinetic energies E_1 and E_2 of the individual photoelectron within a pair. For each $E_{1,2}$, we integrate over the detected angular distribution of photoelectrons. To separate the true DPE signals from a background of accidental coincidence events, a second set of experiments with a 30 times higher photon flux is used. These reference spectra are dominated by the accidental events and serve as the background spectra. The raw and the background spectra are compared in Fig. 1(b) along E_{sum} , with the latter scaled down to the former in the region of $E_{\text{sum}} > 29$ eV, where DPE is prohibited [10]. For clarity, only DPE spectra after this background subtraction are presented subsequently. A DPE cutoff at $E_{\text{sum}}^{sp-d} = 18.9$ eV can be clearly seen in Fig. 1(b), which represents photoemission of the most energetic correlated electron pairs in Ag.

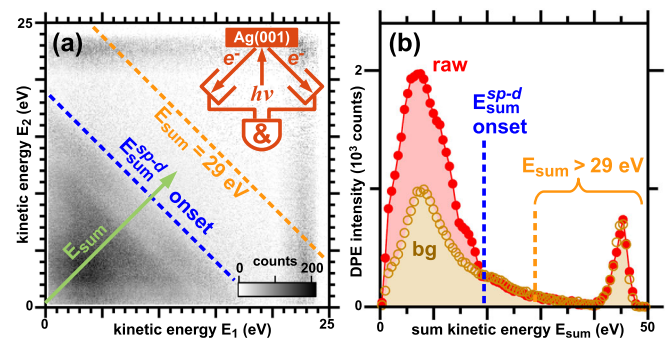


FIG. 1. (a) Histogram of two-electron photoemission coincidence events (raw data) on Ag(001) at $h\nu = 32.3$ eV. The setup is shown schematically in the inset. (b) Raw and scaled background (bg) E_{sum} spectra.

The detailed energy hierarchy of correlated electron pairs is revealed in Figs. 2(a) and 2(b), which are derived from the raw data in Fig. 1 [11]. Below E_{sum}^{sp-d} , we observe two stepwise intensity increases at 14.7 (E_{sum}^{d-d}) and 9.3 eV ($E_{\text{sum}}^{d-d-d^*}$). Moreover, the DPE spectrum in Fig. 2(c) at $h\nu = 25.1$ eV shows two steps at $E_{\text{sum}}^{sp-d} = 12.0$ eV and $E_{\text{sum}}^{d-d} = 7.3$ eV. Since both $h\nu - E_{\text{sum}}^{sp-d}$ and $h\nu - E_{\text{sum}}^{d-d}$ are $h\nu$ independent within ± 0.5 eV [12], we assign E_{sum}^{sp-d} and E_{sum}^{d-d} as well as $E_{\text{sum}}^{d-d-d^*}$ to features of occupied two-electron states that can be specified only by their total energy as a proper quantum number of a two-particle system.

As shown in Figs. 2(b) and 2(c), these two-electron features are located at pair binding energies $E_{B,\text{sum}} = 4.6, 8.8, \text{ and } 14.2 \pm 0.7$ eV. These values are compatible with 1, 2, and 3 times the minimum binding energy of electrons within the Ag $4d$ bands of 4 eV [14]. Therefore, we attribute the first two energies to the onset of pair emission with $sp-d$ and $d-d$ electron-electron assignments, respectively. As an example, we illustrate in Fig. 2(d) the $sp-d$ pair emission process with one sp electron from the Fermi-level (E_F) and one d electron from the top of the d bands. This $sp-d$ process results in the pairs with a maximum kinetic energy of E_{sum}^{sp-d} and is indicated by the dashed line in Figs. 2(c). In analogy, the emission of two electrons from the d bands has a maximum energy of E_{sum}^{d-d} (dashed-dotted line).

Moreover, the spectra in Figs. 2(a) and 2(b) have an intensity increase below $E_{\text{sum}}^{d-d-d^*}$, which corresponds to about 3 times the minimal binding energy of the Ag d bands. Therefore, we explain the marked area $d-d-d^*$ as a lower bound for the emission process of a $d-d$ pair accompanied by another d electron excited up to E_F (d^*) as shown in Fig. 2(d). The labeled $d-d-d^*$ intensity amounts to 40% of the $d-d$ pairs, one order of magnitude larger than conventional shake-up satellites [15], and it overlays on a sizable background [16]. Both these properties are consistent with the known low-energy electron pairs in coincidence Auger spectra on metals, which result from decay processes with at least three valence electrons [17,18]. Because of the characteristic onset $E_{\text{sum}}^{d-d-d^*}$, it is possible to identify the $d-d-d^*$ pairs from other underlying multielectron events. Here we also tentatively exclude $d-d$ pairs with an atomically localized two-hole final state (d^{-2}), since they would contribute a sharp spectrum with a narrow width of about 1 eV [19]. An alternative explanation for $d-d-d^*$ pairs may be the excitation of transient excitons during the $d-d$ pair emission [20,21].

In Fig. 3(a), we compare the DPE spectra of Ag with the self-convoluted density of states (cDOS, dashed line) [22], which is scaled to match the $d-d$ pair intensity. The cDOS gives an estimation for the two-electron DOS as a function of the binding energy of the pairs ($E_{B,\text{sum}}$) and is derived

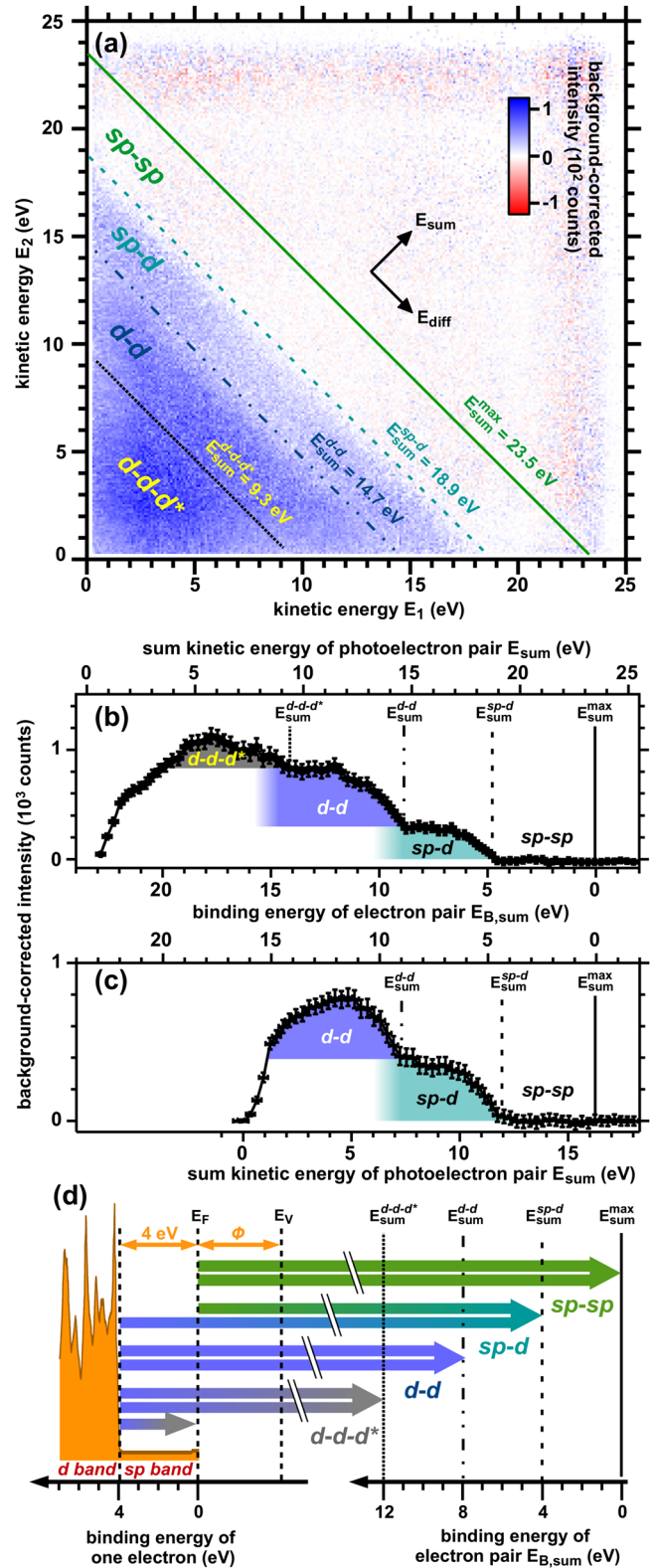


FIG. 2. (a) DPE data at $h\nu = 32.3$ eV on Ag(001). (b) E_{sum} spectrum from (a) integrated over $E_{\text{diff}} = \pm 1$ eV. (c) The same as (b) for $h\nu = 25.1$ eV. (d) DOS of Ag [13] with DPE processes. E_V labels the vacuum level.

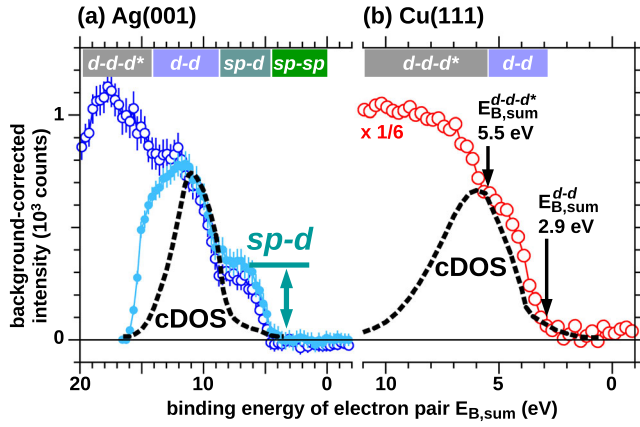


FIG. 3. Comparison of DPE spectra with the self-convoluted DOS adapted from Powell (cDOS) [22]. The constituents of electron pairs are labeled near the upper scale. (a) DPE spectra of Ag(001) at $h\nu = 32.3(25.1)$ eV with empty (solid) symbols from Figs. 2(b) and 2(c). The threefold higher intensity of $sp-d$ pairs compared to cDOS is highlighted. (b) DPE spectrum of Cu(111) with $h\nu = 32.3$ eV integrated over all E_{diff} for better statistics.

from the Cini-Sawatzky model for two valence holes at the weak correlation limit [23,24]. As one can see, the clear DPE feature from $sp-d$ pairs of Ag is surprising. First of all, its intensity amounts to a factor of 3 higher as compared to the cDOS at $E_{B,\text{sum}} = 5-8$ eV. Second, with more than 20% variation in $h\nu$, this enhancement exists persistently. Therefore, significant $h\nu$ -dependent final state effects acting only on $sp-d$ or $d-d$ pairs can be excluded. Possible candidates for such effects are pair diffraction and shake-off [25,26]. Furthermore, identical experiments on Cu(111) give the DPE spectrum in Fig. 3(b) in comparison with the cDOS. There, the $sp-d$ pairs are merely visible, despite the $d-d$ and $d-d-d^*$ features that can be identified similarly as for Ag [27]. To explain all these observations qualitatively, either the two-particle DOS of $sp-d$ electron pairs in Ag must be enhanced relative to the $d-d$ pairs due to correlation, or the DPE matrix element for the $sp-d$ pairs must be significantly larger than for the $d-d$ pairs. Since the DPE matrix element is also linked to the strength of correlation [28–30], our results provide in either case an indication for significant electron correlation between the $5sp$ and $4d$ electrons in Ag.

Since an *ab initio* DPE calculation for Ag does not exist yet, we discuss qualitatively two aspects regarding the $sp-d$ and $d-d$ pairs. The first one is the existence of an electron correlation between sp and d electrons that is stronger in Ag than in Cu. The $sp-d$ electron-electron interaction in transition metals has been postulated to explain itinerant magnetism [31] and the Kondo effect [32]. This interaction is strengthened in solids due the compression of the extended sp wave function towards the d electrons in atomic cores [33]. As a consequence of the larger volume fraction occupied by the $4d$ electrons in comparison to the $3d$ electrons [34], the $sp-d$ interaction can be stronger in

Ag than in Cu, therefore explaining our observation of $sp-d$ pairs in DPE only on Ag. Additionally, this stronger $sp-d$ interaction in Ag also leads to a larger $sp-d$ hybridization at E_F [35]. Moreover, the $\text{Ag}^+ 4d^{10}$ shell has a significantly higher polarizability than the $\text{Cu}^+ 3d^{10}$ shell [36]. As a result, the surrounding $5sp$ electrons in Ag could influence the $4d$ electrons more actively and lead to a significant DPE of $5sp-4d$ electron pairs. This perspective is furthermore consistent with previous band structure calculations, where the different impact of electron correlation on sp electrons in Ag as compared to Cu was implied [37].

A second aspect is the lower strength of electron correlation between the d electrons in the detected $d-d$ pairs than that between the sp and d electrons. Since the on-site Coulomb interaction $U_{\text{on site}}$ between d electrons is usually large up to several eV [38,39], only interacting d electrons at different atomic sites may have a lower strength of correlation than that between the sp and d electrons near the same atom ($U_{\text{intersite}} < 0.5U_{\text{on site}}$ for Ag [40]). This consideration suggests that the observed $d-d$ pairs are a result of the interatomic correlation and leave two photoholes at separated atomic sites ($d^{-1} + d^{-1}$). These d holes can propagate through the lattice with energy dispersion and give rise to a width in the E_{sum} spectrum comparable with the cDOS. It is, however, not straightforward to separate according to DPE spectra the on-site from the band correlation effects, with the latter conventionally assigned to a shift between experimental and theoretical band energies.

In the DPE spectra of both Ag and Cu in Fig. 3, we observe a generally dominant intensity of the $d-d$ and $d-d-d^*$ electron pairs. In strong contrast, vanishing DPE intensity is observed for pairs with both electrons from the bulk sp bands or from the sp -derived Shockley surface state [41]. The observation of electron pair emission as soon as d bands are considered clearly suggests that a band-resolved picture is required for electron correlation, in general, even for metals like Ag and Cu. The vanishing intensity of $sp-sp$ pair is attributed to their low cDOS (Fig. 3) and the less dominant role of electron correlation in the electron gas of the sp electrons [42].

A closer look at Fig. 3(b) reveals a small but observable energy shift between the E_{sum} spectrum and the cDOS for Cu for $d-d$ pairs. However, for Ag in Fig. 3(a), they coincide reasonably. We ascribe this difference to the influence of the stronger Coulomb interaction between d electrons in Cu than in Ag [38,39], which contributes to the repulsion between the two photoholes in the DPE final state and triggers the relaxation of the whole electronic system [43,44]. Because the itinerant nature of correlated valence electrons is unavoidably related to a site-dependent Coulomb interaction [19,45], the atomic models conventionally used for core-valence Auger decays may not be directly applicable to describe the shifted onset observed in our DPE experiments. Therefore, we consider the

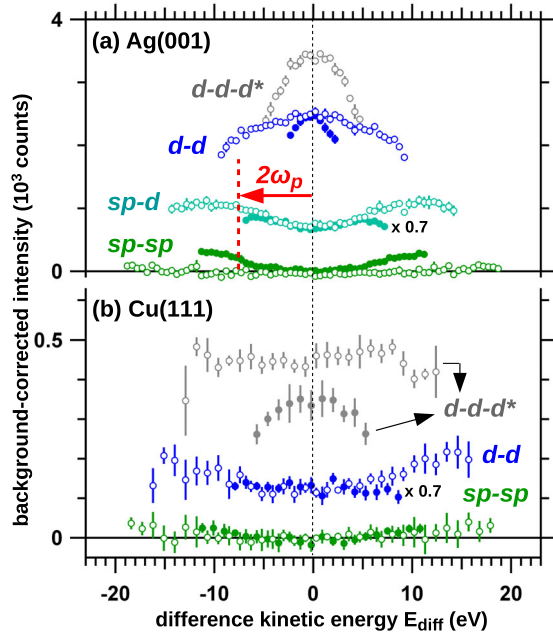


FIG. 4. E_{diff} spectra for (a) Ag(001) and (b) Cu(111) with empty (solid) symbols for $h\nu = 32.3(25.1)$ eV. Each spectrum is integrated over $E_{B,\text{sum}} = \pm 2$ eV for Ag and ± 1 eV for Cu centered at the indicated pair binding energy region in Figs. 2 and 3(b). The $d-d-d^*$ spectra are centered at $E_{B,\text{sum}} - 2$ eV, and the Cu $sp-sp$ spectra at $E_{B,\text{sum}}$ around 1.4 eV. For comparison, the $sp-d$ pairs from Ag and the $d-d$ pairs from Cu at $h\nu = 25.1$ eV are scaled by a factor of 0.7.

difference between the observed Cu $d-d$ onset from the cDOS as an upper bound for the correlation energy of the valence bands DPE process [46], which awaits theories including a refined exchange correlation for a quantitative comparison [33].

The pairs of interacting valence electrons not only have band-specific total energies but also share the exciting photon energy in distinct ways. This can be clearly quantified in terms of the experimentally accessible, kinetic energy difference (E_{diff}), which corresponds to the intensity distribution perpendicular to E_{sum} direction in the DPE histogram as shown in Fig. 2(a). In Fig. 4, the E_{diff} spectra are shown, classified according to the pair configurations discussed above. The E_{diff} spectra for Ag $d-d$ and $d-d-d^*$ pairs show higher intensity at $E_{\text{diff}} \approx 0$. In contrast, $sp-d$ pairs from Ag as well as $d-d$ pairs from Cu are less favorable around $E_{\text{diff}} \approx 0$ when compared to $|E_{\text{diff}}| \gg 0$. These results show a clear partition of $h\nu$ over the two interacting electrons within a pair and its dependence on their valence band character. Moreover, going from $d-d$ to $d-d-d^*$ pairs measured at the same $h\nu$, we observe a more enhanced signal near $E_{\text{diff}} \approx 0$ comparing to larger $|E_{\text{diff}}|$ for both Ag and Cu, which reveals the influence of the additional d^* excitation to the energy distribution within the $d-d$ pairs.

A closer look at the E_{diff} spectrum of $sp-d$ pairs of Ag shows a roughly constant intensity beyond $|E_{\text{diff}}| \geq 2\hbar\omega_p$, with $\hbar\omega_p \approx 3.8$ eV as the bulk Ag plasmon energy [47]. This observation implies that the electrons with otherwise equal energy in an $sp-d$ pair may *additionally* exchange energy via a plasmon and end up as a pair with a larger $|E_{\text{diff}}|$. In contrast, the observed intensity of Cu $d-d$ pairs increases up to a much higher $|E_{\text{diff}}| \approx 10$ eV. This value is comparable to a broad plasmonlike energy-loss resonance [47] as well as to the on-site Coulomb repulsion $U_{\text{on site}} \approx 9-11$ eV [48,49]. The former is excluded because an energy-loss process also alters the total energy (E_{sum}) of an electron pair. Therefore, we attribute the latter as a possible origin for the increasing DPE intensity at larger $|E_{\text{diff}}|$ for Cu $d-d$ pairs, which involves an additional DPE pathway via a reconfiguration of the d shell related to $U_{\text{on site}}$.

In summary, DPE spectroscopy resolves band-specific electron pairs in Ag(001) and Cu(111). In spite of the isoelectronic single-particle band structure of Ag and Cu, they show remarkably different response in the two-particle spectrum. We identify distinct two-particle energy features in the pair sum kinetic energies and relate the energy thresholds according to the number of d electrons involved. Besides electron pairs consisting of $sp-d$ and $d-d$ electrons, we provide indications for processes with three d electrons. The emission of $sp-d$ electron pairs is enhanced on Ag whereas barely observable on Cu due to the weaker $sp-d$ interaction. Moreover, the energy sharing within a pair depends sensitively on the constituent valence electrons and provides hints of energy exchange between electrons via a plasmon in Ag or via the on-site Coulomb interaction in Cu. Our results reveal a clear band dependence in the pairs of mutually interacting electrons in solids, which may pave a way to systematically analyze quasiparticles by multidimensional photoelectron spectroscopy [50].

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