Entangled and Disentangled Decoherence of Intermediate Electron-Hole Pairs in Two-Photon Photoemission from Surface Bands: Beyond the Adiabatic Approximation

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We discuss the effects which combinations of entangled and disentangled decoherence and transient response induce on the early decay of coherent electron-hole pairs in the intermediate states of two-photon photoemission (2PPE) from image potential bands. We find that their interplay gives rise to deviations of the pair decay probabilities from simple exponential laws governed by independent quasiparticle lifetimes obtained in the self-energes based on the adiabatic hypothesis. Assessment of these effects for paradigmatic Cu(111) surface shows that they are most pronounced in the interval of pump-probe photon pulse delay times typical of current 2PPE experiments.

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The development of two-photon photoemission (2PPE) spectroscopy in the 1980s and 1990s has greatly facilitated the studies of linewidths [1,2] and lifetimes [3–6] of quasiparticle states of the surface image potential [7,8]. In 2PPE from a surface state (SS) band the photons from the first, pump pulse may excite electrons from occupied states into intermediate unoccupied states below the vacuum level E_V. Photons from the second, probe pulse may excite these electrons into evanescent wave states above E_V so that the resulting 2PPE yield is governed by quadratic response of the system to the applied photon field [9,10]. If the intermediate states of the experiment lie within the image potential (IP) band, the photoemitted electrons provide *combined* information on the evolution of IP and SS states that proceeds through two distinct stages. In the former, between the absorption of pump and probe pulse photons, the system evolves through a *neutral excited* state in which the optically generated IP-SS electron-hole (*e-h*) pair is as a *whole* subjected to dynamical interactions with the rest of the system. In the latter, upon emission of the IP electron by the probe photon, the system is left in *ionized final* state with a relaxing and diffusing SS hole.

One of the main goals in the time resolved 2PPE experiments is the determination of (de)coherence of excited electronic states of the investigated systems. The evolution of coherent IP-SS *e-h* pairs in the intermediate states of 2PPE, which convey relevant information on the properties of excited states, is affected by *disentangled* and *entangled* decoherence and dephasing caused by the coupling of the pair constituents to the heatbath of the system and by their scattering off impurities. Dominant contribution to disentangled decoherence arises from individual interactions of the optically excited electron or hole with the electronic charge density fluctuations (Auger type of transitions) in the substrate. These processes can be quantified in terms of renormalizations of the corresponding *one-particle propagators* in the time domain from which the single quasiparticle decay and transition probabilities are calculated. The quasiparticle lifetime is then usually identified with the inverse linewidth in the spectrum of quasiparticle propagator in the energy domain. By assuming adiabatic switching on of the interaction with the heatbath, the linewidth is related to the imaginary part of the quasiparticle self-energy Σ that in lowest order perturbation theory is obtained from Fermi's golden rule (FGR). Entangled decoherence arises from the mutual interaction of the optically excited electron and hole in the intermediate states and is also dominantly mediated by the substrate electronic density fluctuations. This type of decoherence is described by the vertex renormalization that accounts for excitonic effects in the corresponding *e-h pair propagator*.

The Σ-approach calculations of lifetimes of single quasiparticles in the surface bands, based on the adiabatic hypothesis, linear response formalism, and the various levels of approximations ranging from FGR to GW, were performed for a number of metal surfaces [11,12] and compared with the photoemission, inverse photoemission, scanning tunneling microscopy and 2PPE data. The time scale of applicability of adiabatic assumption in the evaluations of individual quasiparticle lifetimes was subsequently examined for a paradigmatic Cu(111) surface with partly occupied SS and the unoccupied IP band [13,14]. It was found that the regime of exponential single quasiparticle decay with the lifetime obtained in the Σ approach is established few femtoseconds after the primary excitation of a quasiparticle. Hence, past that interval in which the quasiparticle picture cannot be firmly established [15], the Σ approach is expected to reliably describe *single* quasiparticle decay processes, with the nonadiabatic corrections in the form of Debye-Waller factors for the decay probabilities.

Besides the range of applicability of adiabatic description for quasiparticle evolution on the early time scale there still remains a question of the neglected role of entangled decoherence [16], which is a prerequisite for making quantitative comparisons between the calculated and 2PPE-deduced quasiparticle lifetimes. In this Letter we assess for the first time the relative roles of entangled and disentangled decoherence and characteristic time scales of intermediate IP-SS e-h pairs for the same prototype surface Cu(111) for which the applicability of the Σ approach was examined [14]. We start from a simple two-band model described in [13,14] that proved convenient in studying the substrate response to transient perturbations and making contact with the calculations in the Σ approach [17–21]. To assess how the dominant elementary processes affect the entangled decoherence we introduce several simplifying assumptions for the propagation of an optically excited IP-SS e-h pair. First, we ignore interband diffusion and interband transitions that give rise to the decay of quasiparticles out of the bands in which they were generated by the pump pulse [19] because such processes do not much affect the importance of entangled decoherence on the short time scale we aim to explore. Second, as we are interested in the propagation of a hole excited near the SS-band bottom, whose average recoil energy in the excitation of substrate quanta is much smaller than the occupied SS-band width, we also neglect all exchange effects involving this hole and holes excited near the Fermi level. With these approximations the optically excited hole can diffuse (recoil) across the occupied part of SS band in one time direction only [22,23].

In the absence of decoherence each component of the once created coherent IP-SS e-h pair state evolves independently. Hence, a measure of decoherence of the pair at time $t > 0$ is deduced from transition amplitudes [15]

$$a_{\mathbf{K}',\mathbf{K},n}(t) = \langle \mathbf{K}', n | U(t) c^{\dagger}_{\mathbf{K},\mathrm{IP}} c_{\mathbf{K},\mathrm{SS}} | 0 \rangle, \tag{1}$$

where $|0\rangle$ is the substrate ground state, $c^{\dagger}_{\mathbf{K},\mathrm{IP}}$ and $c_{\mathbf{K},\mathrm{SS}}$ are the creation and annihilation operators for electrons with momentum $\hbar\mathbf{K}$ in the quasi–two-dimensional IP and SS bands, respectively, and $U(t)$ is the evolution operator of the system. The initial IP-SS state at $t = 0$ is given by $c^{\dagger}_{\mathbf{K},\mathrm{IP}} c_{\mathbf{K},\mathrm{SS}}|0\rangle = |\mathbf{K}, 0\rangle$, and it is assumed that the momentum supplied by the pump photon is negligible. The state $|\mathbf{K}', n\rangle$ can be any intermediate state of the system in which IP electron and SS hole propagate with momenta $\hbar\mathbf{K}'$ and $-\hbar\mathbf{K}'$, respectively, and n real quanta of the substrate charge density fluctuations are excited in the substrate. The Hamiltonian H of the present two-band model [14] embodies the coupling of IP-electron and SS-hole charge to the charge density fluctuations in the substrate whose dynamics is described by the electronic response function. We also assume translational invariance along the surface and quadratic dispersion of IP- and SS-quasiparticle energies characterized by the effective masses m_{IP} [2,24] and m_{SS} [24], respectively.

Most relevant to the assessment of the relative importance of entangled and disentangled decoherence effects and their relation to the Σ-approach results are the amplitudes $a_{\mathbf{K}',\mathbf{K},n}(t)$ of transitions that are dominant on the early time scale. These are the transitions with zero ($n = 0$) and one real quantum ($n = 1$) of the substrate charge density fluctuations present in the system at instant t (see below). For $n = 0$ there are two physically distinct contributions: the amplitude of diagonal transition, $R^{(\mathrm{diag})}_{\mathbf{K}}(t) = a_{\mathbf{K},\mathbf{K},n=0}(t)$, whose absolute square

$$P^{(\mathrm{diag})}_{\mathbf{K},n=0}(t) = |a_{\mathbf{K},\mathbf{K},n=0}(t)|^2 \tag{2}$$

yields the decay probability of the initial IP-SS state in the course of time, and the sum of amplitudes of off-diagonal transitions, $R^{(\mathrm{offd})}_{\mathbf{K}}(t) = \sum_{\mathbf{K}'} a_{\mathbf{K}',\mathbf{K},n=0}(t)$, which describes the scattering of IP-SS e-h pair through the exchange of substrate quanta. The probability

$$P^{(\mathrm{offd})}_{\mathbf{K},n=0}(t) = |\sum_{\mathbf{K}'} a_{\mathbf{K}',\mathbf{K},n=0}(t)|^2 \tag{3}$$

is a direct measure of the entangled decoherence of the optically excited IP-SS e-h pair.

For $n = 1$ only the off-diagonal amplitudes are nonvanishing and the sum

$$P^{(\mathrm{offd})}_{\mathbf{K},n=1}(t) = \sum_{\mathbf{K}'} |a_{\mathbf{K}',\mathbf{K},n=1}(t)|^2 \tag{4}$$

yields the total probability of finding an IP-SS e-h pair scattered out of the initial state, with one excited quantum of the substrate charge density fluctuations carrying away the momentum $\hbar(\mathbf{K} - \mathbf{K}')$. Lowest order perturbation contributions to this sum calculated in the adiabataic limit are proportional to the transition rates $\Gamma^{\mathrm{IP}}_{\mathbf{K}}$ and $\Gamma^{\mathrm{SS}}_{\mathbf{K}}$ that describe, respectively, the IP-electron and SS-hole state decay caused by disentangled decoherence.

In the calculations of transition probabilities (2), (3), and (4) under the assumptions on hole dynamics described above we resort to the exponentiated Born approximation formalism [25], which produces very accurate results in comparison with exact numerical solutions [26]. Figure 1 shows $P^{(\mathrm{IP-diag})}_{\mathbf{K}=0,n=0}(t)$ and $P^{(\mathrm{SS-diag})}_{\mathbf{K}=0,n=0}(t)$ which describe independent decays of initial IP-electron and SS-hole states on Cu(111) surface caused solely by disentangled decoherence processes. The results illustrate a particularly instructive case of optical transitions at the $\bar{\Gamma}$ point ($\mathbf{K} = 0$) because in transitions between the bottoms of SS and IP bands there is no available phase space for downward intraband recoil in IP-electron interactions with the heatbath. Hence, $\Gamma^{\mathrm{IP}}_{\mathbf{K}=0} = 0$ in the absence of interband transitions as available decay channels. This causes a nonexponential decay of the electron state at the IP-band bottom and leads to a saturation of its decay probability at the value of IP Debye-Waller factor (DWF),

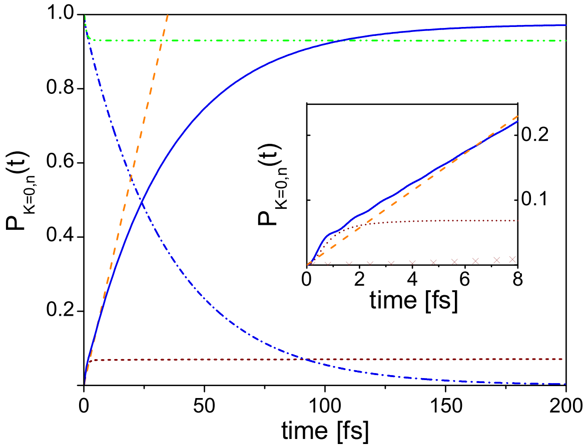

FIG. 1 (color online). Single particle decay probabilities induced by disentangled decoherence on Cu(111) surface for SS hole, $P^{(\mathrm{SS-diag})}_{\mathbf{K}=0,n=0}$ (dash-dotted line), and IP electron $P^{(\mathrm{IP-diag})}_{\mathbf{K}=0,n=0}$ (upper dash-dot-dotted line). Total transition probabilities for SS hole $P^{(\mathrm{SS-offd})}_{\mathbf{K}=0,n=1}$ (solid line), and IP electron $P^{(\mathrm{IP-offd})}_{\mathbf{K}=0,n=1}$ (lower dotted line) into a final excited state involving a single substrate excitation quantum. The dashed line denotes SS-hole FGR transition probability $\propto t$. Inset: Comparison of the early evolution of one-quantum $P_{\mathbf{K}=0,n=1}$ and FGR (lines), and two-quantum $P^{(\mathrm{SS-offd})}_{\mathbf{K}=0,n=2}$ (crosses) transition probabilities.

$$P^{(\mathrm{IP-diag})}_{\mathbf{K}=0,n=0}(t \to \infty) = \exp(-2W^{\mathrm{IP}}_{\mathbf{K}=0}) = \mathrm{const} < 1, \quad (5)$$

which measures the weight of elastic line in the relaxed IP-electron spectrum. In contrast, for $\mathbf{K} = 0$ the recoil of SS hole can take place across the whole occupied part of SS band of width $B = 0.39$ eV [24], but the average recoil energy is $\epsilon \sim 30$ meV $\ll B$. This gives rise to nonzero $\Gamma^{\mathrm{SS}}_{\mathbf{K}=0}$ and to the asymptotic behavior

$$P^{(\mathrm{SS-diag})}_{\mathbf{K}=0,n=0}(t \gg \hbar/\Gamma^{\mathrm{SS}}_{\mathbf{K}=0}) = \exp(-2W^{\mathrm{SS}}_{\mathbf{K}=0}) \exp(-\Gamma^{\mathrm{SS}}_{\mathbf{K}=0} t). \quad (6)$$

Here $\exp(-2W^{\mathrm{SS}}_{\mathbf{K}})$ is the SS-hole DWF that measures the weight of elastic line in the relaxed SS-hole spectrum.

In Fig. 1 we also show the magnitudes of fundamental off-diagonal transition probabilities $P^{(\mathrm{IP-offd})}_{\mathbf{K}=0,n=1}(t)$ and $P^{(\mathrm{SS-offd})}_{\mathbf{K}=0,n=1}(t)$ for the single IP electron and SS hole, respectively, which include all possible uncorrelated emissions and reabsorptions of the substrate quanta in renormalizations of the initial quasiparticle propagators, and the final emission of one real quantum propagating at instant t. Quite generally, $P^{(\mathrm{IP-offd})}_{\mathbf{K},n=1}(t)$ and $P^{(\mathrm{SS-offd})}_{\mathbf{K},n=1}(t)$ start as $\sim t^2$, which is a general characteristic of the short time response to transient perturbations, with non-Markovian oscillations of $P^{(\mathrm{SS-offd})}_{\mathbf{K},n=1}(t)$ clearly discernible in the first few fs (see inset). However, for longer times they may display a very different behavior, depending on the initial quasiparticle position in its band. For $\mathbf{K} = 0$ we have $0 < P^{(\mathrm{IP-offd})}_{\mathbf{K}=0,n=1}(t \to \infty) \ll 1$, whereas $P^{(\mathrm{SS-offd})}_{\mathbf{K}=0,n=1}(t)$ for $t \gg \hbar/\Gamma^{\mathrm{SS}}_{\mathbf{K}=0}$ approaches much closer to unity in its deviation from the adiabatic FGR-like transition probability $\propto t$ that is also displayed for comparison. In the absence of entangled decoherence the independent transition probabilities satisfy unitarity conditions $P^{(\mathrm{IP-diag})}_{\mathbf{K},n=0}(t) + \sum_{n=1}^{\infty} P^{(\mathrm{IP-offd})}_{\mathbf{K},n}(t) = 1$ and $P^{(\mathrm{SS-diag})}_{\mathbf{K},n=0}(t) + \sum_{n=1}^{\infty} P^{(\mathrm{SS-offd})}_{\mathbf{K},n}(t) = 1$. Here, owing to the DWF's close to unity, these unitarity sums are for $t < \hbar/\Gamma^{\mathrm{SS}}$ almost completely exhausted by the transitions with $n = 0$ and $n = 1$.

In Fig. 2 we show the diagonal and off-diagonal IP-SS *e-h pair* transition probabilities $P^{(\mathrm{diag})}_{\mathbf{K}=0,n=0}(t)$ and $P^{(\mathrm{offd})}_{\mathbf{K}=0,n=0}(t)$, respectively, calculated in the presence of entangled decoherence and transient effects under the same assumptions as for the quantities in Fig. 1. Also shown for comparison is the corresponding adiabatic exponential decay probability for IP-SS *e-h* pair that is free from these corrections. The off-diagonal transition probability $P^{(\mathrm{offd})}_{\mathbf{K}=0,n=0}(t)$, which is nonzero only in the presence of entangled decoherence, starts as $\sim t^4$ in the very short initial interval $\tau_{\mathrm{IS}} \simeq (v_F Q_{\mathrm{IS}})^{-1}$ where v_F is the substrate Fermi velocity and $\hbar Q_{\mathrm{IS}}$ is the maximum momentum exchange in entangled decoherence processes involving a single substrate excitation quantum. $P^{\mathrm{offd}}_{\mathbf{K}=0,n=0}(t)$ may reach a maximum provided $\hbar/(\Gamma^{\mathrm{SS}} + \Gamma^{\mathrm{IP}}) > \tau_\mu$ (present case) where $\tau_\mu = 2\mu/(\hbar Q^2_{\mathrm{IS}})$ is the differential recoil induced decoherence time with $\mu = |1/m_{\mathrm{SS}} - 1/m_{\mathrm{IP}}|^{-1}$. After this it falls off as t^{-1} to the asymptotic limit. In the opposite case $\hbar/(\Gamma^{\mathrm{SS}} + \Gamma^{\mathrm{IP}}) < \tau_\mu$ it uniformly rises towards the asymptotic limit only

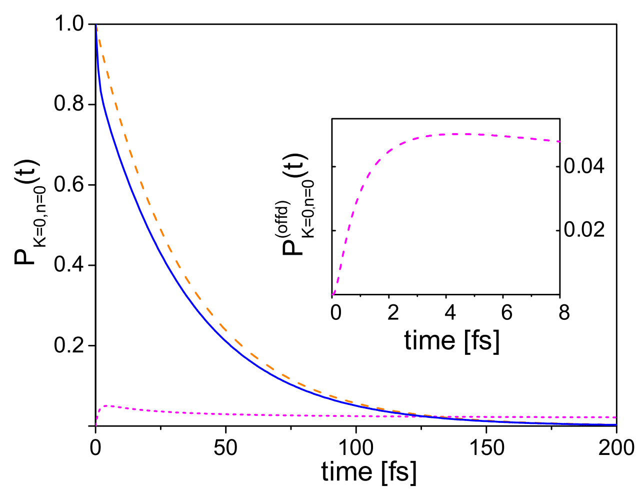

FIG. 2 (color online). Diagonal IP-SS electron-hole pair decay probabilities on Cu(111) surface calculated in the presence (full line) and absence (dashed line) of entangled decoherence and transient effects, for initial $\mathbf{K} = 0$. Dotted line is the total off-diagonal entangled decoherence induced transition probability $P^{(\mathrm{offd})}_{\mathbf{K}=0,n=0}$. Inset shows $P^{(\mathrm{offd})}_{\mathbf{K}=0,n=0}$ in the early time interval.

weakly dependent on $(\Gamma^{\rm SS}+\Gamma^{\rm IP})$. Hence, the entangled decoherence probability is governed by an interplay between single quasiparticle and pair propagations and its magnitude is affected by $\Gamma^{\rm SS}$, $\Gamma^{\rm IP}$, and τ_μ. This affects the total decay probability of the IP-SS *e*-*h* pair as a whole, as is seen by comparing the bare exponential decay derived in the Σ approach with $P^{\rm (diag)}_{\mathbf{K}=0,n=0}(t)$ that embodies entangled decoherence and transient effects (Fig. 2). In the presence of all these effects the total IP-SS pair decay probability contains three contributions to the total DWF and for $\mathbf{K}=0$ asymptotically behaves as

$$P^{\rm (diag)}_{\mathbf{K}=0,n=0}(t\gg\hbar/\Gamma^{\rm SS}_{\mathbf{K}=0})=\exp(-2W^{\rm SS}_{\mathbf{K}=0})\exp(-2W^{\rm IP}_{\mathbf{K}=0})\times\exp(-2W^{\rm IS}_{\mathbf{K}=0})\exp(-\Gamma^{\rm SS}_{\mathbf{K}=0}t), \tag{7}$$

where $\exp(-2W^{\rm IS}_{\mathbf{K}=0})$ is the entangled decoherence induced DWF. The first three exponentials on the right-hand side of (7) represent nonadiabatic and entangled decoherence corrections to the bare exponential decay described by the last exponential. The largest correction to the bare exponential decay occurs in the interval <50 fs that is typical of pump-probe pulse delay times in current 2PPE experiments. For $\mathbf{K}\neq 0$ the decay rate $\Gamma^{\rm IP}_{\mathbf{K}}$ in (5) and (7) is nonzero, and in the presence of interband transitions both $\Gamma^{\rm SS}_{\mathbf{K}}$ and $\Gamma^{\rm IP}_{\mathbf{K}}$ are additionally modified [19,27]. However, in both situations the general structure of expression (7) persists, with total $\Gamma_{\mathbf{K}}=\Gamma^{\rm IP}_{\mathbf{K}}+\Gamma^{\rm SS}_{\mathbf{K}}$.

For long times $P^{\rm (SS-offd)}_{\mathbf{K},n=1}(t)$ and $P^{\rm (IP-offd)}_{\mathbf{K},n=1}(t)$ are of the order $\mathcal{O}(\lambda_{\rm SS})$ and $\mathcal{O}(\lambda_{\rm IP})$, respectively, where $\lambda_{\rm SS(IP)}=Q_{\rm SS(IP)}\Omega_{\rm SS(IP)}\partial^2 S_{\mathbf{Q}}(\omega)/\partial Q\partial\omega|_{Q=0,\omega=0}$. Here $\hbar Q_{\rm SS(IP)}$ and $\hbar\Omega_{\rm SS(IP)}$ denote the maximum momentum and energy transfer to a substrate quantum excited by the SS hole (IP electron), respectively, and $S_{\mathbf{Q}}(\omega)$ is the substrate excitation spectrum [14]. On the other hand, $P^{\rm (offd)}_{\mathbf{K},n=0}(t)=\mathcal{O}((\lambda_{\rm IS}\tau_\mu/\tau_{\rm IS})^2)\propto\mu^2$, and thus for large μ or $m_{\rm IP}\sim m_{\rm SS}$ (resonant enatanglement) it can be of the same order of magnitude as $P^{\rm (IP-offd)}_{\mathbf{K}=0,n=1}(t)$. Conversely, the absence of dispersion in the intermediate states [as, e.g., in the antibonding states of Cs/Cu(111) [28]] renders $\mu=m_{\rm SS}$, which precludes resonant scattering and hence weakens the excitonic interaction in the case of *e*-*h* pairs optically excited at the $\bar{\Gamma}$ point. If $\lambda_{\rm SS}\sim\lambda_{\rm IP}\sim\lambda_{\rm IS}$ the entangled and disentangled decoherence effects become equally important and tend to largely cancel out each other in the $\mathbf{K}$-resolved IP-SS polarization propagator $R_{\mathbf{K}}(t)=-i\Theta(t)(R^{\rm (diag)}_{\mathbf{K}}(t)+R^{\rm (offd)}_{\mathbf{K}}(t))$ (c.f. [14]).

In summary, we have demonstrated that in 2PPE from surface bands on Cu(111) the intermediate state decays are affected by entangled decoherence and transient effects to the amount that necessitates their inclusion in the analyses and comparisons of quasiparticle lifetimes obtained from experiment and theory. This calls for assessments of these effects in other systems for which the Σ-approach single particle lifetimes are already available.

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