Femtosecond decoherence of quasiparticles in the states of surface image potential:

Beyond the lifetime effects given by Fermi's golden rule

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We discuss the femtosecond dynamics of quasiparticles (electrons and holes) in the states of image potential at metal surfaces that can be probed and investigated by various spectroscopic techniques. The occupied electronic states of surface state bands (SS-bands) are commonly investigated by photoemission spectroscopy (PES) in which absorption of a photon induces ejection of an electron from the system, leaving a hole in one of the previously occupied SS-band states. The suddenly created SS-hole interacts with the screening charge in the system and this dynamic interaction causes its decoherence that can be traced over several time scales, each of which gives rise to specific effects in the measured photoelectron spectra. A complementary technique of inverse photoemission spectroscopy (IPES or BIS), in which electrons are injected into initially unoccupied band states, can probe the dynamics of electrons in unoccupied image potential bands (IP-bands). The injected electrons are subjected to a dynamical interaction with the system which gives rise to their decoherence in a fashion analogous to the case of holes created in PES from occupied states, with similar effects concerning the various time scales. In time resolved two-photon photoemission spectroscopy (2PPES) applied to probe the occupied states of SS-bands by the pump pulse and the unoccupied states of IP-band by the probe pulse, both type of transient interactions will affect the photoelectron yield. Sudden creation of a surface localized electron-hole (e-h) pair that evolves into an IP-SS e-h pair in the first step of 2PPE effectively switches on the same type of transient interactions which the holes and electrons are individually subjected to in PES and IPES, but in addition to these there also appears a mutual IP-SS e-h interaction that gives rise to interference effects. Due to the delay time between the pump and probe pulses, which is desirably in the femtosecond range, it is of special interest to investigate as how the IP electron, the SS hole, and the IP-SS e-h pair as a whole evolve on that time scale. In particular, it is important to assess at which instant there occurs a crossover between the propagation of quasiparticles that is characterized by the pre-exponential decay caused by transient interactions, and the exponential decay in which the decoherence effects can be described in terms of quasiparticle lifetimes. The latter are obtainable from Fermi's golden rule and have been widely used to interpret the lifetime aspects of PES, IPES and 2PPES measurements.

In this work we illustrate and quantify the various decoherence effects on the femtosecond time scale for the case of propagation of electrons, holes and optically created electron-hole pairs in 2PPE from two dimensional IP- and SS-bands on the paradigmatic Cu(111) surface[1]. We show how different types of temporal evolution of quasiparticles appear on different time scales and point out at which stage Fermi's golden rule approach for calculating lifetimes of quasiparticles becomes applicable to describe the decoherence effects in 2PPE from this prototype system.

[1] B. Gumhalter, Surf. Sci. 518(2002)81.