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Coherent polarization dynamics in three-photon photoemission from Cu(111) surface CONG WANG, XUEFENG CUI, SEAN GARRETT-ROE, HRVOJE PETEK, Univ of Pittsburgh — We investigate the surface states on Cu(111) by multi-photon photoemission using tunable ultra-short (<15 fs) laser pulses. The angle-resolved photoemission spectra in the pre-resonant region for the two-photon excitation from the Shockley surface (SS) state to the n=1 image potential state are consistent with the well-known band structures. An anomaly is observed, however, for excitation at 610 nm (2.04 eV), where the tripling of the SS band occurs, the SS appearing as three parallel bands with the same effective mass. Excitation with the doubled frequency does not support this phenomenon. Instead we attribute this to parallel excitation pathways where the multi-photon photoemission occurs in response to the external and internal fields. The external field at 2.04 eV can drive the multiphoton absorption from SS, but also the coherent local field associated with the transition from top of the d-band to the sp-band at its crossing with the Fermi level can drive the photoemission. We examine the coherent nonlinear polarization dynamics by performing interferometric two-pulse correlation measurements. Fourier transform with respect to delay axis provides 2D linear and nonlinear spectra of the coherent polarization leading to the observed phenomena.

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