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Ultrafast plasmon-enhanced hot electron process in model heterojunctions: Ag/TiO_2 and Ag/graphite[1]

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We study the plasmonically enhanced nonlinear photoemission from Ag nanocluster-decorated graphite and $TiO_2(110)$ surfaces by time-resolved two-photon photoemission spectroscopy (TR-2PP). Evaporating Ag atoms on graphite and TiO_2 surfaces forms pancake-like Ag clusters with 5 nm diameter and 1-1.5 nm height through self-limiting growth mode. The Ag nanoparticles enhance the two-photon photoemission (2PP) signal by approximately two-orders of magnitude as compared with the bare surfaces for p-polarized excitation. In the case of s-polarization there is essentially no enhancement for graphite, and only about an order-of-magnitude enhancement for TiO_2. Wavelength dependent measurements of the enhancement reveal that for Ag/graphite there is a single plasmonic resonance due to the $\perp$-plasmon mode at 3.6 eV. By contrast, for Ag/TiO_2 there are $\perp$ and $||$-plasmon modes with resonant energies of 3.8 and 3.1 eV, respectively. Apparently the dielectric properties of the substrate have strong influence on the type and frequency of Ag plasmonic modes that can exist on the surfaces. 2PP spectra of the Ag/graphite and Ag/TiO_2 surfaces reveal two distinct components that are common to both. The high energy component consists of a coherent 2PP process from an occupied interface state, which only exists in the presence of Ag. We identify this state, as an interface state formed by charge donation from the Ag-5s band to the unoccupied states of the substrates. The low energy component consists of a hot electron signal that is created by plasmon dephasing. TR-2PP measurements are performed on the plasmon-induced electron dynamics to assess their relevance for plasmonically enhanced femtochemistry.

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