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Ultrafast Hot Carrier Scattering and Generation from Surface Plasmons in Noble Metals MARCO BERNARDI, JAMAL MUSTAFA, Department of Physics, University of California, Berkeley, and Materials Science Division, Lawrence Berkeley National Lab, Berkeley, CA 94720, USA, JEFFREY B. NEATON, Molecular Foundry, Lawrence Berkeley National Lab, Berkeley, CA 94720, USA, STEVEN G. LOUIE, Department of Physics, University of California, Berkeley, and Materials Science Division, Lawrence Berkeley National Lab, Berkeley, CA 94720, USA — Non-equilibrium "hot"carriers in materials are challenging to study experimentally as they thermalize at subpicosecond time and nanometer length scale. Recent experiments employed hot carriers generated by light absorption or surface plasmon annihilation in noble metals (e.g., Au and Ag) for catalysis and solar cells. The energy distribution and transport of the generated hot carriers play a key role in these experiments. We present ab initio calculations of the energy distribution of hot carriers generated by surface plasmons in noble metals, and the relaxation time and mean free path of the hot carriers along different crystal directions within 5 eV of the Fermi energy. Our calculations show the interplay of the noble metal s and d bands in determining the damping rate of the plasmon and the mean free path of the hot carriers. The trends we find as a function of surface plasmon momentum and frequency allow us to define optimal experimental conditions for hot carrier generation and extraction. Our approach combines density functional theory, GW, and electron-phonon calculations. Our work provides microscopic insight into hot carriers in noble metals, and their ultrafast dynamics in the presence of surface plasmons.

Marco Bernardi
Department of Physics, University of California, Berkeley

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