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Broadband Hot Electron Creation in Gap Plasmon Nanostructures¹ GARY WIEDERRECHT, MATTHEW SYKES, Argonne National Laboratory, JON STEWART, GLEB AK-SELROD, Duke University, DAVID GOSZTOLA, Argonne National Laboratory, XIANG-TIAN KONG, University of Electronic Science and Technology of China, MAIKEN MIKKELSEN, Duke University, ALEXANDER GOVOROV, Ohio University — Plasmonic metallic nanostructures typically exhibit narrow resonances that can be tuned from the visible to infrared through geometry changes. Upon photoexcitation, plasmons rapidly decay to produce hot electrons near the metal's surface through either intra- or interband transitions. These charge carriers initially form a nonthermal energy distribution. Here we demonstrate the spectral and temporal response of both nonthermal and thermal hot electrons excited in a metasurface comprising gold or silver nanoparticles separated from a plasmonic thin film with a dielectric spacer of nanometric thickness. The ultrafast spectral response from hot electrons is shown to extend over 1000 nm from the ultraviolet to near-infrared wavelengths. Through experiment and modeling, we describe how such populations couple to the various optical frequencies and metasurface modes, and the response is shown to occur efficiently down to very low fluence. The extreme broadband and ultrafast response offers improved understanding of nonthermal systems far from equilibrium.

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