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Electron and hole dynamics in the electronic and structural phase transitions of VO_2[1]

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The ultrafast, optically induced insulator-to-metal transition (IMT) and the associated structural phase transition (SPT) in vanadium dioxide (VO_2) have been studied for over a decade. However, only recently have effects due to the combined presence of electron-hole pairs and injected electrons been observed. Here we compare and contrast IMT dynamics when both hot electrons and optically excited electron-hole pairs are involved, in (1) thin films of VO_2 overlaid by a thin gold foil, in which hot electrons are generated by 1.5 eV photons absorbed in the foil and accelerated through the VO_2 by an applied electric field; (2) VO_2 nanoparticles covered with a sparse mesh of gold nanoparticles averaging 20-30 nm in diameter in which hot electrons are generated by resonant excitation and decay of the localized surface plasmon; and (3) bare VO_2 thin films excited by intense near-single-cycle THz pulses. In the first case, the IMT is driven by excitation of the bulk gold plasmon, and the SPT appears on a few-picosecond time scale. In the second case, density-functional calculations indicate that above a critical carrier density, the addition of a single electron to a 27-unit supercell drives the catastrophic collapse of the coherent phonon associated with, and leading to, the SPT. In the third case, sub-bandgap-energy photons (approximately 0.1 eV) initiate the IMT, but exhibit the same sub-100 femtosecond switching time and coherent phonon dynamics as observed when the IMT is initiated by 1.5 eV photons. This suggests that the underlying mechanism must be quite different, possibly THz-field induced interband tunneling of spatially separated electron-hole pairs. The implications of these findings for ultrafast switching in opto-electronic devices – such as hybrid VO_2 silicon ring resonators – are briefly considered.

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