

Dissipative Dynamics of Excited States in Nanointerfaces

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Silicon is one of the most technologically important materials with innumerable applications in microelectronics and photovoltaics. To realize the full potential of nanostructured Si for photovoltaic devices, it is necessary to explore the photoinduced electron transfer pathways. Metal adsorbates such as Ag clusters introduce electronic states in the energy gap of Si and provide stronger absorption in the near-IR to visible spectral region, opening up additional pathways for surface charge transfer.^{1,2} In this work, we study charge carrier dynamics in atomistic models composed of Ag clusters adsorbed on Si surface. Specifically, such a nanointerface is modeled under photoexcitation by a light pulse or continuous wave irradiation. The relaxation of the photoinduced hot carriers is modeled by an account of the electron-phonon interactions treated by the on-the-fly nonadiabatic couplings between electronic and nuclear degrees of freedom. The nonadiabatic couplings serve as parameters for the reduced density matrix approach within the Redfield formalism. Our results show that the relaxation rates of hot electrons are higher than that of hot holes due to more relaxation channels. By comparison with pure Si surface, we find that the introduction of the adsorbates leads to a favorable pattern of charge carrier relaxation and stronger intense light absorption, which is advantageous for applications to photovoltaics and photocatalysis.

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References:

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