Nonlinear excitation of solids and transient band gap dynamics upon femtosecond laser irradiation of semiconductors: insights from first principles simulations

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The irradiation of bandgap materials by intense and ultrashort laser pulses leads to the excitation of numerous quantum and macroscopic phenomena [1-3]. To describe a wide variety of physical and chemical processes, both first-principle theories and phenomenological descriptions are of importance [4]. In particular, time-dependent density functional theory (TDDFT) is an efficient first-principle method to predict the excitation dynamics of electrons in laser-irradiated band-gap materials.

In this work, the transient excitation of silicon was studied by extensive TDDFT simulations. The density of conduction band electrons, the total electronic current and the electronic energy were calculated for a wide range laser intensities and for different laser wavelengths. The approach has enabled to achieve a detailed description of the transition from the multiphoton to the tunneling regime of photoionization, characterized by a decrease of a number of photons required for excitation of valence electrons to the conduction bands. The associated nonlinear absorption rates were calculated as a function of laser intensity for several wavelengths. It was found that, at high laser intensities, the density of the excited conduction electrons saturates to the number of electrons initially present in the valence band while, after saturation, free electrons continue to absorb laser energy. The dependence of the free-electron energy on wavelength has peaks, which shift with increasing the laser field strength.

To explain these effects, the dynamics of the electron energy levels in the laser field was studied using a Floquet Hamiltonian, which describes the influence of the laser field on the electronic bands. At low laser intensities, the replicas of the electronic bands are found to locate at energies shifted in comparison to the ground state that is explained by the interaction between the electronic bands. When intensity is increasing, the replicas can cross, manifesting the transient closure of the band gap. This effect is related to the electron tunneling and can be described by the dynamic Floquet-Stark metallization [5-7].

As a whole, this work provides quantitative information that is of high importance for the improvements of theories describing excitation, metallization and damage of laser-irradiated bandgap materials, an important step for gaining more control over ultrashort laser processing of semiconductors and dielectrics.

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