Simulation of energy flow in femtosecond-laser excited h-BN/graphene heterostructures

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Recent years show increasing interest in 2D vdW materials after unique properties as well as new effects in those materials were observed. Phenomena linked to heterostructures are, but not limited to, interlayer excitons, new phonon modes, band structure hybridization or Moiré superlattice effects such as unconventional superconductivity. A promising way to induce but also investigate new processes in 2D vdW heterostructures is femtosecond-laser pulses because of their pulse duration comparable to typical time scales of atomic motions in molecules and solids. Additionally, the ability to shape amplitude, phase, and polarization within one pulse provides a variety of excitation channels. More general, the excitation of a crystalline material by an intense ultrashort-laser pulse creates extreme nonequilibrium conditions within the solid. The optical energy of such an excitation is, due to the high difference in mass of electrons and atoms, mainly deposited in the electronic system. Therefore one can induce electronic temperatures in materials of several 10000's of K, whereas the atomic system remains almost unaffected near room temperature. Such an immense change in the electronic system has a direct influence on the bonding properties of the material and is the origin of several ultrafast phenomena. But how is the deposited energy distributed after the excitation within the material and on which timescale? The answer to this question becomes even harder in the case of 2D vdW heterostructures. Why? Compared to, e.g., silicon inter-material correlations of electrons and phonons in vdW heterostructures, like h-BN/graphene, open interaction and energy diffusion channels that might not be present in either of the single materials. Recent research showed, e.g., out-of-plane heat transfer by electron-hyperbolic phonon coupling or ultrafast relaxation of hot phonons. In order to investigate the strength and direction of energy flow within and between heterostucture layers after a femtosecond-laser excitation we performed ab initio calculations of mono-layer hBN-graphene using the real-space and real-time density-functional-theory (DFT) code Octopus. The laser-pulse excitation is simulated by an time-dependent vector potential with a Gaussian shaped envelope. The now time-dependent response of the electron density is computed with time-dependent DFT (TDDFT). The characterizing parameters of the corresponding electrical field are the amplitude A = 0.072836 eV/Å that corresponds to a peak intensity of 7.04×10^{10} W/cm^2, the wavelength 800 nm corresponds to $\hbar\omega$ = 1.549 eV, and the pulse duration t = 25 fs. The timestep for the electronic evolution is t = 0.33 as. The induced ionic motion by the changing electronic system is computed in accordance to Ehrenfest dynamics. Note that hBN is transparent for the used wavelength of 800 nm: it does not absorb laser-pulse energy, whereas graphene/graphite does. In this setup we assure that only the electronic system of carbon-based subsystem is excited so that the ensuing energy flow into different channels can be identified and quantified. Our findings allow us to give a theoretical picture on the thermalization process within both materials but also between them. Moreover, by comparing our simulation results to experimentally obtained time-resolved ultrafast-electron-diffraction data we are enabled to verify our theoretical predictions but also improve our theoretical approach to a point where reliable prediction to thermal and non-thermal contributions and their corresponding timescales can be obtained. Those results are an important step towards understanding of inter-material correlations which can be used to control the ensuing structural response.