Ultrafast field control of symmetry and reversibility in buckled graphene-like materials

Hamed Koochaki Kelardeh, **Vadym Apalkov**, Mark Stockman

Center for Nano-Optics (CeNO) and Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303, USA

vapalkov@gsu.edu

We study theoretically interaction of buckled two-dimensional graphene-like materials (silicene and germanene) with a femtosecond (one optical oscillation) strong optical pulse with the amplitude $\sim 1\ V/$Å. We show that such graphene-like materials subjected to such strong optical field can be controlled by the optical field component normal to their plane. This is due to specific buckled structure of silicene/germanene, which results in sensitivity of both the bandgap and the interband dipole coupling to the normal component of the optical field. Microscopically, the normal component of the strong field causes transfer of electrons between two sublattices of graphene-like materials, resulting in decreases of the symmetry of the system from hexagonal (six-order, centrosymmetric) to triangular (third-order, non-centrosymmetric).

We show numerically that in strong ultrashort optical fields, the buckled graphene-like materials exhibit non-reciprocal reflection, optical rectification and generation of electric currents both parallel and normal to the in-plane field direction. Reversibility of electron dynamics in the field of the pulse is also field- and carrier-envelope phase controllable. Here we characterize the reversibility of electron dynamics by the residual conduction band population, i.e. conduction band population after the pulse. Namely, the dynamics is reversible if, after the pulse ends, the conduction band population, which is the residual conduction band population, is small compared to the maximum conduction band population throughout the pulse. We show that by variation of the angle of incidence and the amplitude of the pulse, the electron dynamics can be changed from highly irreversible, where the maximum conduction band population is attained at the end of the excitation pulse, to partially reversible: at the end of the pulse the conduction band population is reduced by a factor of ≈ 2 with respect to its maximum.

There is a net charge transfer along the material plane that is also dependent on the normal field component. The effective reduction of symmetry of the buckled graphene-like materials from hexagonal to triangular (where there is no inversion center), caused by the strong normal field component of the pulse, causes the currents in the silicene lattice to be highly anisotropic and non-reciprocal. The temporal dependence of the current shows loss of adiabaticity in higher fields, which also implies irreversibility and, consequently, violation of time-reversal symmetry (T-symmetry). One of the consequences of the T-invariance violation are non-zero values of the transferred charge and of the residual polarization. This also implies that the system's dynamics is irreversible (non-adiabatic), which may surprise one because the system is completely Hamiltonian. This is due to the fact that the central frequency of the laser radiation, $\approx$ 1.5 eV, is close to the transition frequency between the electron states localized at the two sublattices of graphene-like materials. This causes resonant absorption leading to dephasing - collisionless relaxation widely known as Landau damping.

The currents, generated in the graphene-like materials during the pulse, cause transfer of charge across the system and accumulation of charges by the end of the pulse. The charge transferred depends on the field amplitude. A remarkable property of this dependence is that the charge transferred changes its sign as the field amplitude increases. This can be attributed to the increased number of electrons experiencing the Bragg reflections at the Brillouin zone boundary with the field increase. The charge accumulated at the pulse end is an experimentally observable quantity just as in the previous experiments on currents in dielectrics [1].

We note a close analogy of silicene/germanene with the field-effect transistor (FET). In FET, the gate field, applied normally to the conducting channel, changes the carrier populations in it and, thereby, controls its conductance. Analogously, in silicene/germanene, the normal field component, transfers carriers to one of the sublattices, thereby changing the system's response to the in-plane field. A fundamental difference (and advantage) of silicene/germanene is that such a ``device'' works at optical frequencies, with the response time on the (sub)femtosecond scale. This opens a potential for many applications of silicene in future Petahertz-speed devices and applications.

References

[1] A. Schiffrin, *et al.*, Nature, **493** (2012) 70.