Nonlinear optics and transport of excitons in two-dimensional crystals

M.M. Glazov
Ioffe Institute, 194021,St.-Petersburg, Russia
glazov@coherent.ioffe.ru

Two-dimensional transition metal dichalcogenides form a family of direct-band semiconductors with strong excitonic effects. Due to relatively large effective masses of the charge carriers and rather weak screening of the Coulomb interaction Wannier-Mott excitons with the binding energies on the order of several hundreds of milli-electronvolts dominate the optical properties of these exemplary two-dimensional materials. Excitons form a series of 1*s*, 2*s*, 2*p*, etc., hydrogen-like states. The exciton wavefunctions and binding energies deviate from the two-dimensional hydrogenic series mainly due to specific screening of the Coulomb interaction in two-dimensional systems.

Here we present the results of theoretical and experimental investigations of several nonlinear optical and transport effects on excitons in MX_2 monolayers, where M is the transition metal and X is the chalcogen (M=Mo,W and X=Se,Se as a rule).

First, we address two-photon absorption by excitons. We show that both *s*- and *p*-excitons are active in two-photon processes. This is because MX_2 monolayers lack an inversion center. The symmetry of the monolayers allows for the mixing of the *s*- and *p*-excitons. Moreover, it allows linear in the wavevector terms in the interband transition matrix element. Both effects contribute to the two-photon activity of *s*-excitons. By contrast, the two-photon absorption on *p*-excitons does not require any symmetry reduction [1].

Due to simultaneous single and two-photon activity of excitons, the second harmonic generation becomes possible. In this nonlinear process a given state is excited by the two photons and emits coherently a single photon of a double frequency. We demonstrate the second harmonic emission it is strongly enhanced if the fundamental frequency is twice smaller than the exciton resonance frequency. The mechanisms of the second harmonic are established [1].

Furthermore, we study the exciton-mediated upconversion, i.e., the two-photon absorption where the single photon energy is close to the exciton energy. Due to efficient Auger process one of the photocreated excitons recombines nonradiatively transferring its energy to another one. It gives rise to the upconversion photoluminescence [2].

Finally, we study excitonic transport in two-dimensional MX_2 semiconductors. Depending on the exciton density three regimes of exciton transport can be identified. First, in the linear regime exciton-exciton interactions are unimportant and the excitons simply diffuse in the monolayer plane. Second, with an increase in the exciton density the Auger recombination comes into play making exciton distribution in the real space more flat and giving rise to an increase of the apparent diffusion coefficient. Third, for even higher densities the memory effects become important and a halo-like distribution of excitons in the real space is formed with a dip at the excitation spot [3].

[1] M. M. Glazov, L. E. Golub, G. Wang, X. Marie, T. Amand, and B. Urbaszek, Phys. Rev. B **95**, 035311 (2017).
[2] M. Manca, M. M. Glazov, C. Robert, F. Cadiz, T. Taniguchi, K. Watanabe, E. Courtade, T. Amand, P. Renucci, X. Marie, G. Wang, B. Urbaszek, Nature Communications **8**, 14927 (2017).
[3] A. Chernikov, M.M. Glazov, et al., Nonlinear exciton propagation and halo effects in atomically thin semiconductors, in press.