“Excitonic design of perovskite LEDs and lasers”

This project aims to design hybrid organic-inorganic (or inorganic) perovskites, 3D and low-dimensional, for chosen ranges of the excitonic parameters, which are suitable for lasers (with very strong exciton binding energy E_b), LEDs (moderate E_b), and (...not intentionally) photovoltaics (very low E_b). Second focus is on the excitonic life-times for fast optoelectronics in communication technology. Third goal, most ambitious, concerns the electrically-driven laser, since the perovskite-community faced serious difficulties with its realization, despite advances with the optically-pumped lasers.

The organic-inorganic lead halide perovskites (and lead-free perovskites) have formula ABX_3, where A=Cs,Rb,methylammonium(MA),formamidinium(FA), B=Pb,Sn, and X=Cl,Br,I. Perovskites are ionic crystals, where A^+ and B^{2+} are cations and X^- is an anion. In light-emitting devices (LEDs), the perovskite layer is used as an efficient recombination center. It is sandwiched between the electron transporting layer (ETL, n-type) and hole transporting layer (HTL, p-type). Lasers need mirrors (a cavity) in order to achieve the amplified spontaneous emission. However, perovskites have large refractive indexes at air-interface (2.2-2.55) and small critical angles, enabling light amplification without a cavity if low-dimensional structures are used. Confinement is also advantageous for the desired high exciton-binding energies (thus brightness) and low treshold current for an optical gain. Many LEDs and optically-pumped lasers have been published since 2014. Both types pulse and continuous-wave lasers are available, in addition to the first-field application in the solar cells, where the power-conversion efficiency increased from 3.8% in 2009 up to 23.7% in 2018 (silicon PCE=26.6%).

The bandgap is mainly ruled by a choice of halide ion: $MAPbCl_3$ and $MAPbBr_3$ have a direct bandgap of 2.97 eV (417 nm) and 2.23 eV (556 nm), respectively, and $MAPbI_3$ has slightly indirect bandgap of 1.55-1.7 eV (730-800 nm). Larger bandgaps of 3.3 eV (376 nm) are available in double perovskites ABi(Ag,Cu)X6, and much smaller bandgap, <0.5 eV (>2480 nm), is in Li_3SPbI_3. Mixing perovskites not only enables to obtain all intermediate bandgaps (laser color), but also efficiently increases the stability of the devices. Additionally, the halide-ion exchange can be used as a way of introducing confinement, thus, increasing the radiative recombination rate and busting up the excitonic energies. To date, the perovskite materials and their nanostructures offer a wide range of the electron-hole coupling of 43-376 meV, and this is not the last word. Our aim is to extend the above borders.

The workplan includes the chemical (doping) and structural (low-dimensions) modifications designed in such a way that the excitonic radius (the electron-hole localization) is small and the exciton binding is strong. Moreover, we will shift the energy levels of the A-cation with respect to the levels of the BX_3 “frame”, expecting that this will allow a charge injection into the molecule, for the purpose of electrically-pumped laser. Although cavities are not necessary for nanostructures, the polariton laser (with strong exciton-photon coupling) should be easier to construct if one applies good mirrors, where “good” means the topological edge-states. This idea seems to be promising for perovskites, due to the fact that both topological edge-states and Dirac-cone band structures were found in doped or compressed perovskites, or in their 2D layers cut along some special crystal directions.

The theoretical methods, planned to be used in this project, are based on the density-functional theory (DFT) and many-body perturbation theory (beyond DFT). First type of calculations sets an input for the second, which include the electron-screening effects due to absorption of light. With such tools, one is able to get such parameters as: exciton binding energy and radius, true optical spectra, exciton life-time and current treshold for the optical gain. In addition to the band structure, which is an usual outcome from the DFT.