Revealing up-conversion mechanisms in PbI_2 crystals

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In typical situation, the photoluminescence of a given material occurs at energy which is lower than the energy of the absorbed photons. Opposite case (i.e. anti-Stokes type emission) is less common, as it requires pooling together the energy of several absorbed photons. As such, *up-converting* materials are of principal interest for photovoltaic and other energy harvesting applications.

In this work we focus on one of materials exhibiting effect of up-conversion: lead(II) iodide (PbI_2). This material has layered structure, which exhibits transition from direct to indirect band gap when crystal is thinned to monolayer. Crystals of PbI_2 exhibit very efficient luminescence about 500 nm even under excitation at longer wavelength.

Here we report initial studies of low-temperature photoluminescence (PL) of PbI_2 crystals as a function of excitation power and excitation energy. Our measurements reveal presence of multiple regimes, and consequently - multiple up-conversion mechanisms. The CW excitation results in linear dependence of up-converted photoluminescence intensity on the excitation power, which can occur, e.g., in case of full saturation of intermediate levels. On the other hand, the excitation with femtosecond laser leads to quadratic or higher power dependence. Such dependence is usually a strong indication of a mechanism based on two-photon absorption, yet it is only partially supported by the measurements of up-conversion efficiency as a function of excitation energy, in particular in the range corresponding to half of band gap energy. In our work we discuss possible reasons behind this disparity and analyze the cross-over regime between CW and pulsed mode.