Band structure and exciton dynamics in mono and multi-layer 2D perovskites

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Quasi two-dimensional perovskites in Ruddlesden-Popper phase (RPPs) are extensively studied for their flexible tunability of the optoelectronic properties by changing the number of layers, chemical composition and the organic spacer, which makes them particularly suitable for several applications in the field of photovoltaics and light emitters. In this developing framework, the photoexcited carrier dynamics is still far to be completely understood although the study of charge generation and recombination path can furnish key information for boosting the development of new materials for new generation optoelectronic. Here we present time-resolved measurements performed with transient absorption spectroscopy of the RPP DAMAPI ($DA_2MA_{l-1}PbI_{3l+1}$, where $DA=CH_3\text{-}(CH_2)_{11}\text{-}NH_3^+$ is the spacer and l indicate the number of the layers) performed with the excitation pump at selected energies and at 77 and 300 K. These measurements allowed us to obtain both spectral and temporal information about excitonic absorption and decay. We show that the derivative-like shape of the transient absorption signal is very sensitive to small changes of the carrier population, allowing us to observe electronic transition well above the band gap energy. Beyond the observation of the n=1 exciton, we have been able to observe in DAMAPI a clear absorption bleaching related to the excitons with principal quantum number n=2. The interpretation of the data has been supported by theoretical calculations of the band structure of DAMAPI based on BSE and GW model using the YAMBO platform. The analysis of the decay of the transient absorption signal has pointed out the dominant contribution of the exciton-exciton annihilation mechanism in quenching the excitonic absorption bleaching.