EPJ Web of Conferences **238**, 07006 (2020) https://doi.org/10.1051/epjconf/202023807006
EOSAM 2020

Tuning the transient opto-electronic properties of few-layer MoS_2 nanosheets via substrate nano-patterning

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Abstract. We study the optical properties of Molybdenum Disulphide nanosheets deposited by chemical vapor deposition onto a nanopatterned substrate endowed with a uniaxial corrugation. The uniaxial nanocorrugation leads to a polarization dependence in steady state and an anisotropic relaxation around the C resonance. Finite element numerical simulations allow to better disclose the scenario. Our findings point towards new possibilities for the manipulation of the optical properties of two-dimensional transition metal dichalcogenides via substrate nanopatterning.

Nowadays two-dimensional transition metal dichalcogenides (TMDs) constitute one of the most promising class of materials for novel opto-electronic devices due to their peculiar physical properties including strong light-matter interaction and excitonic effects [1]. As recently demonstrated, their properties can be modified via substrate pattern design by introducing an anisotropic degree of freedom in the spatial arrangement of TMD nanosheets [2, 3].

In this context, a new scalable deposition methodology which allows chemical vapour deposition growth over macroscopic area (cm^2 range) of Molybdenum Disulfide (MoS_2) has been applied to a self-organized nano-patterned substrate with sub-wavelength rippled shaped corrugation [4].

So far, the effect of nano-patterning on the opto-electronic properties of TMDs has been considered only in terms of Raman modes, photoluminescence, absorption whereas the effects of morphology induced anisotropy in the time domain has been largely overlooked.

In this work, we present a detailed analysis of the linear and the transient optical properties of a few layer MoS_2 nanosheets, deposited on top of a SiO_2 substrate endowed with a uniaxial nano-corrugation [5]. Our study of the morphology dependent behavior is based on polarization resolved optical extinction and broadband ultrafast pump-probe experiments with 100fs temporal resolution. The experimental evidences, supported by full-wave finite element numerical simulations, show a rich and complex scenario. This is characterized by: (i) a giant optical anisotropy in the spectral region of the four main excitonic resonances (A, B, C, D) with peculiar features dictated by of the out-of-plane component of MoS_2 dielectric permittivity, (ii) a polarization dependence of the relaxation dynamics around the C resonance (Fig. 1) where the optical response is dominated by a van Hove singularity in the joint density of states. We argue that the polarization dependent dynamics could involve different electron-phonon scattering mechanisms enhanced by the substrate nano-patterning thus allowing the possibility of anisotropy-related exciton engineering.

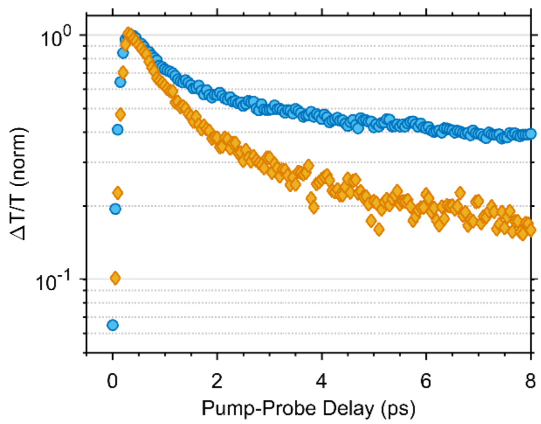

Fig. 1. Normalized transient transmission ($\Delta T/T$) dynamics around the spectral position of the C exciton for different probe polarization direction: parallel (circles) and orthogonal (diamonds) to the corrugation axis.

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The same nano-corrugation of the SiO_2 substrate can also be exploited for the deposition of an array of metal nanowires in close contact with MoS_2 nanosheets [2]. This hybrid metal/semiconductor interface can be exploited for increasing the photon harvesting properties of TMDs and could potentially enable hot carrier based photodetection over cm^2 areas.

Our approach show that substrate nano-patterning is a viable tool for tailoring the opto-electronic properties of two-dimensional materials and provide a suitable platform for new applications in the field of hot carrier technology.

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