Distinguishable separation of charged and neutral excitons at room temperature in anisotropic photoluminescence of TMD monolayers integrated in polymer waveguides



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Abstract

In this work, we propose a suitable integration of WSe2 and MoSe2 in a Poly(methyl methacrylate) (PMMA) in order to properly study the excitation and generation of light of transition metal dichalcogenides (TMD) materials in a waveguide configuration. In particular, we demonstrate that the monolayer (ML) can be homogeneously pumped by the evanescent field of the modes confined in the PMMA, and efficiently excited at room temperature. In addition, the PL emitted by the TMD is not only measured from the surface, but also coupled to the modes of the structure and decoupled at the output edge of the waveguide with an anisotropic behaviour, with a room temperature separation between the TMD photoluminescent components related to the neutral and charged excitons (X⁰ [1] and X^C, respectively). To the best of our knowledge, this is the first time where PL of TMD is properly coupled in a purely horizontal waveguide configuration, and the first approximation to distinguish the X^{C} and X^{0} contributions within the same device at room temperature. Thus, we believe that these results can pave the road of new photonic devices based on TMDs.

Experimental setup



Figure 1. Geometry of the experimental setup and different collection possibilities, as shown in the inset :

- Horizontal excitation Vertical collection
- Horizontal excitation Horizontal collection
- Vertical excitation Vertical collection
- Vertical excitation Horizontal collection

TMDs ML prepared by mechanical exfoliation were transferred through all-dry viscoelastic transfer onto PMMA waveguides on Si/SiO₂ substrates.

Light from a laser was coupled to the edge of the waveguide, and the evanescent field of the confined mode in the PMMA excites the ML, which emits light through PL.

The PL from the TMD ML can be collected from the top of the setup or at the end of the waveguide, collecting the PL that coupled to the waveguide.

Measurements on vertical-vertical were taken on a confocal setup for a micrometric study of the photoluminescent emission.

Simulation analysis



Figure 2. Simulation of the propagation by a multilayer algorithm [3].



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Figure 3. Structure of the samples and electric field distribution of the TE₀ modes at different wavelengths.

The inclusion of the TMD ML barely changes the mode distribution, only a small change ($\approx 0.01\%$) on the effective refractive index of the modes was found. The TMD can be seen homogeneously excited by the evanescent field of the mode that propagates through the waveguide. Part of its PL then couples to the waveguide and also propagates.



Figure 4. a), b) : PL measurements taken in both vertical-vertical and horizontal-vertical configuration for both WSe₂ and MoSe₂. c), d) : Gaussian deconvolution of the PL measurements of horizontal-vertical configuration.

These PL measurements show a spectrum for both materials which clearly change depending on the configuration of the setup. In vertical-vertical configuration, the asymmetry on the neutral exciton peak (X⁰) [4] can be ascribed to the presence of charged excitons (trions, X^c) [5] or localized states. When the measurements were taken in horizontal-vertical configuration, this peak separates into a double-peak, in both materials and all samples studied.

After deconvolution of the horizontal-vertical measurements, two peaks are more clearly recognized. This natural separation due exclusively to the configuration of the setup would allow for a different physical treatment of both components, allowing to tune both separately as desired for a specific application.

Double-peak behaviour



To better understand the double-peak behaviour shown in the horizontal-vertical configuration, measurements in verticalvertical configuration were done. This allowed for some more information on the energy range of the peak associated with charged excitons.

Conclusions

2D MLs of WSe₂ and MoSe₂ have been implemented into a waveguide, exploring not only horizontal excitation – vertical collection, but in all configurations possible. This implementation, specially the horizontal-vertical, shows how 2D materials can be added to integrated optoelectronic devices, as well as current Si integrated technologies. Besides that, two different contributions in the PL emission of TMDs have been observed at RT : the neutral exciton X⁰ and the trion X^c. Being able to separate these contributions in the horizontalvertical configuration allows for further study and interaction without requiring complex conditions such as low temperatures or magnetic fields, which to our knowledge, it has been the first time that this has been achieved. Future works would focus into a better understanding of the intrinsic mechanisms of this separation, and possible applications for it.



Figure 5. a) Optical image from one of the studied samples. Concrete points have been marked. b) PL map of the integrated X^c distribution (1.45-1.52 eV) from the same sample. c) PL measurements from the marked points.

As shown on the fig. 5 a), the point 1, on the center of the sample, shows a typical PL spectra, with its neutral exciton being dominant. When measuring on the borders of the sample, like points 2 or 3, the contribution from the X^c peak grows until being dominant over the neutral exciton due to the high population of localized defects that recombine in positive trions.

This can also be seen at fig. 5 b), an integrated PL map over the energy range of the X^c peak, where the borders show a stronger response.

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