Carrier and polarization dynamics in monolayer MoS_2 studied by time resolved photoluminescence

L. Bouet[1], D. Lagarde[1], X. Marie[1], G. Wang[1], C.R. Zhu[2], B.L. Liu[2], T. Amand[1], P.H. Tan[3], and B. Urbaszek[1]

[1] Toulouse University, INSA-CNRS-UPS, LPCNO, 135 Av. de Rangueil, 31077 Toulouse, France
[2] Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China
[3] State Key Laboratory of Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, Beijing,100083, China

urbaszek@insa-toulouse.fr

Abstract.
Transition metal dichalcogenides such as MoS_2 emerge as an exciting class of atomically flat, two-dimensional materials for electronics, optics and optoelectronics [1]. In contrast to graphene, monolayer (ML) MoS_2 has a direct bandgap in the visible region of the optical spectrum. Inversion symmetry breaking (usually absent in graphene) together with the spin-orbit interaction leads to a unique coupling of carrier spin and k-space valley physics. The circular polarization (σ+ or σ-) of the absorbed or emitted light can be directly associated with selective carrier excitation in one of the two non-equivalent K valleys (K+ or K-, respectively) in momentum space [2]. The chiral optical selection rules open up very exciting possibilities of manipulating carriers in valleys with contrasting Berry phase curvatures, aiming for experimental manifestations of the predicted valley Hall effect [4]. Also stable spin states have been predicted for valence and conduction states for this material.

Up to now optical valley initialization in ML MoS2 is based on the analysis of the large circular polarization degree P_c of the emitted light from the direct bandgap observed in time-integrated measurements following circularly polarized laser excitation [2,5]. An important drawback seemed to be the drastic decrease of P_c as the temperature is raised to 300K [6]. In a simple approach, the time integrated polarisation is determined by the initially created polarization P_0, the lifetime of the electron-hole pair τ and the polarization decay time τ_s through $P_c=P_0/(1+\tau/\tau_s)$. We emphasize that the polarization decay time does not correspond directly to the carrier spin flip time as in most semiconductors like GaAs, but it includes the scattering time between the two non-equivalent K valleys (K+ or K-) [4].

Here we present time resolved polarization measurements in MoS2 monolayers, providing vital information on the valley dynamics from 4K to room temperature [7]. We determine the key parameters that govern the stationary polarization degree P_c: Using quasi-resonant excitation of the A-exciton transitions, we can infer from Figure 1a that the photoluminescence (PL) decays within $\tau \approx 4.5$ps. For pulsed laser excitation, we observe a decrease of P_c with increasing laser power. We show that the PL polarisation remains nearly constant within our time resolution for experiments from 4K up to 300K (see Figure 1b and 1c), a necessary condition for the success of future Valley Hall effect experiments based on optically initialized K-valley polarization [4]. In addition, τ does not vary significantly over this temperature range [7]. These results are surprising when considering the decrease of P_c in time-integrated experiments when going from 4K to 300K reported in the literature [5,6]. By tuning the laser following the shift of the A-exciton resonance with temperature we are able to recover at room temperature 80% of the polarization observed at 4K in our sample, see Figures 2a and 2b. The absence of a clear PL polarization decay within our time resolution suggests that the initially injected polarization P_0, which dominates the steady state PL polarization, is responsible for this observation.

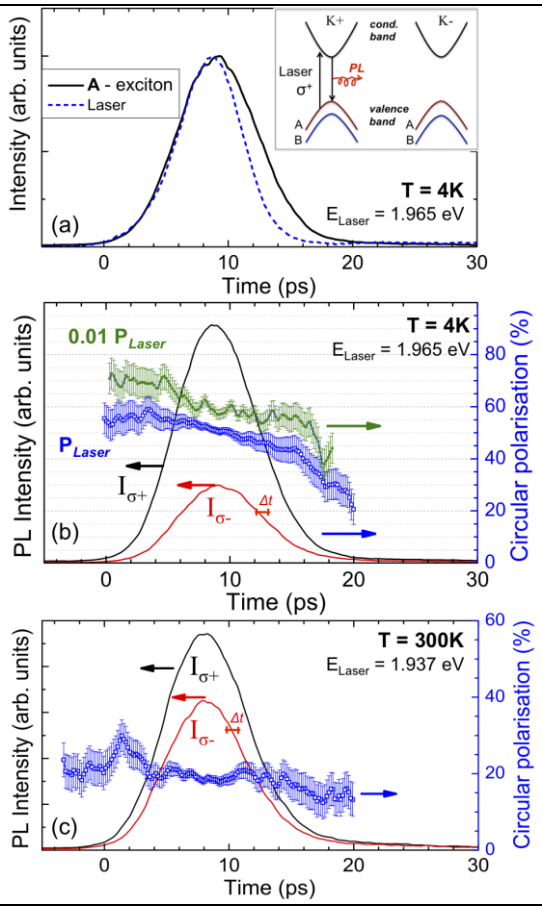
The role of strong excitonic effects (electron-hole pairs bound by Coulomb Interaction) merits further investigation in this context [3], since the broad monolayer PL emission originates from charged and/or neutral exciton recombination. Neutral excitons from the K- and K+ valleys can couple via the Coulomb exchange interaction [8]. The role of this coupling as a mechanism for valley depolarization is discussed.

References

[1] Qing Hua Wang et al, Nat. Nanotechnol, **7** (2013) 699 and references therein
[2] T. Cao et al , Nat. Commun. **3** (2012) 887.
[3] T. Cheiwchanchamnangij et al , PRB **85** (2012) 205302; Diana Y. Qiu et al, PRL **111** (2013) 216805
[4] D. Xiao et al, Phys. Rev. Lett. **108** (2012) 196802
[5] *for example* K. F. Mak, K. He, J. Shan, and T. F. Heinz, Nat. Nanotechnol. **7** (2012) 494.
[6] *for example* G. Sallen, L. Bouet, X. Marie *et al*, Phys. Rev. B. **86** (2013) 081301(R)
[7] D. Lagarde, L. Bouet et al Phys. Rev. Lett. **112**, (2014) 047401.

[8] T. Yu et al, arxiv 1401.0047 and H. Yu et al arxiv 1401.0667

Figure 1



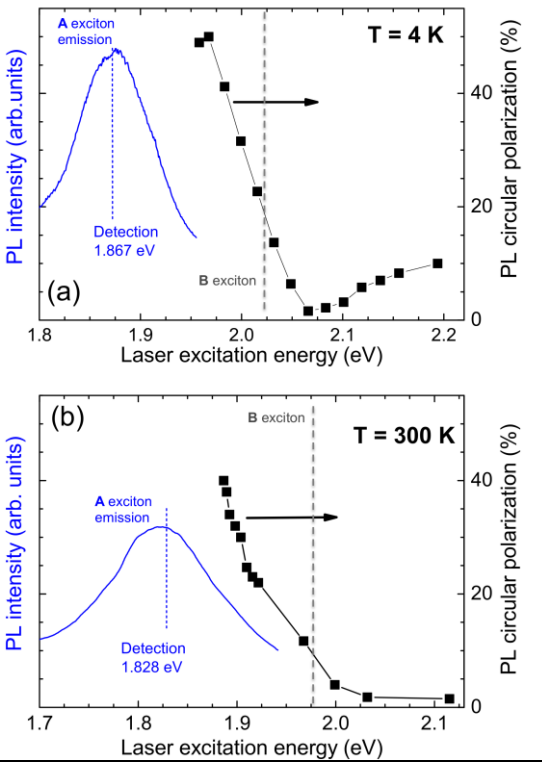
Time resolved photoluminescence of A-exciton in monolayer MoS_2.

(a) Laser pulse (blue line) and PL emission (black line) intensity at T = 4K detected at maximum of A-exciton PL E_{Det}=1.867eV as a function of time. **Inset:** Chiral optical selection rules in 1ML MoS2

(b) T=4K, E_{Laser}=1.965eV, E_{Det}=1.867eV. Laser polarization σ+. Left axis: σ+ (σ-) polarized PL emission intensity presented in black (red) as a function of time. Right axis: Circular polarisation degree during exciton emission.
blue hollow squares: excitation power P_{Laser}=550μW/μm^2,
green full squares:0.01 P_{Laser}, errors bars take into account uncertainty in time origin Δt ~ 0.7ps.

(c) same as (b), but for T=300K, E_{Laser}=1.937eV, E_{Det}=1.828eV.

Figure 2



PL polarization as a function of laser excitation energy in monolayer MoS_2.

(a) T = 4K, P_{Laser}=550μW/μm^2
black squares: PL circular polarization degree detected on the A-exciton PL maximum
E_{Det}=1.867 eV.
Blue spectrum: The A-exciton emission is shown.

(b) same as (a) but for T = 300 K,
P_{Laser}=550μW/μm^2, E_{Det}=1.828 eV