Ab initio exciton – phonon dynamics in Transition Metal Dichalcogenides

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Abstract

One of the most intriguing properties of transition metal dichalcogenides (TMDs) is the topological coupling between spin and valley degrees of freedom. Using a circularly polarised laser, a single spin and valley can be populated, with demonstrated magnetization lifetimes up to microseconds. The carriers scatter between valleys until a balance is reached, but there is significant debate about the relaxation processes for electrons momentum, energy, and spin. The literature debates the relative importance of the electron-electron, electron-phonon, and defect interactions. Time-resolved Kerr rotation measurements on $MoSe_2$ reveal an intriguing temperature dependence of the magnetization sign, which switches as temperature is increased [1]. We explain this behaviour using a fully first-principles many-body approach that explicitly takes into account the carrier-phonon interaction to compute exciton dynamics after laser excitation [2]. In this framework, we show how phonons determine carrier and spin relaxation on a 1-10 ps time scale, as a function of temperature, and how the Kerr signal and total magnetisation behave at different temperatures, explaining the experimentally observed non-monotonic behaviour as a function of temperature [1]. We bring further insight into the remaining mystery of which states are capable of preserving the magnetization on the very long observed time scales.

References

[1] M. Ersfeld, F. Volmer, P. M. M. C. de Melo, R. de Winter, M. Heithoff, Z. Zanolli, Ch. Stampfer, M. J. Verstraete, B. Beschoten, Nano Lett. 19 (2019) 4083

[2] P. M. M. C. de Melo and A. Marini, Phys. Rev. B 93 (2016) 155102

Figures

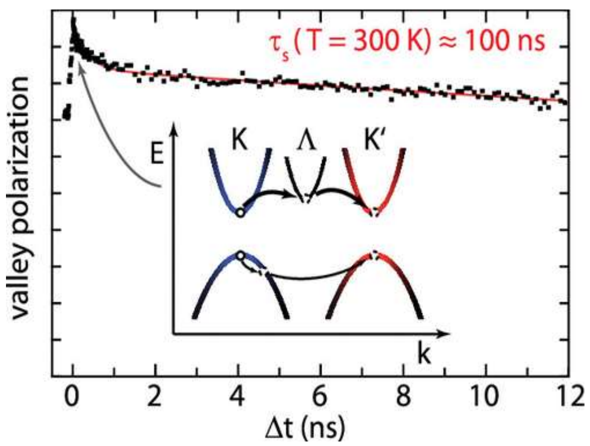

Figure 1: Electron-phonon interaction explains spin relaxation at short timescales [3]