Ultrafast Spin and Charge Transfer in Monolayer WSe_2-Graphene Heterostructure Devices

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Monolayer transition metal dichalcogenides (TMDs) such as WSe_2 have garnered much attention due to their long spin/valley lifetimes and ability to optically excite spin/valley polarization. Additionally, one of the great strengths of TMDs is their ability to compliment other materials, such as graphene, by acting as a means of optical spin injection or proximity coupling. Recently multiple groups have observed efficient, proximity mediated charge transfer in TMD/graphene heterostructures [1]. Moreover, magnetoresistance measurements have demonstrated the ability to optically inject spins from TMD to graphene [2]. However, little work has been done to quantify the speed of spin transfer across a TMD/graphene interface or its effect on spin/valley lifetime.

Here we use time-resolved Kerr rotation (TRKR) microscopy to image the spatial dependence of spin/valley dynamics in monolayer WSe_2/graphene heterostructures. Under p-type gating, Kerr rotation spatial maps of bare CVD grown WSe_2 demonstrate a characteristically spatial dependent, long-lived lived spin/valley signal [3]. Surprisingly though, these Kerr maps show strong quenching of spin/valley density at the WSe_2/graphene interfaces. Time delay scans of the interfaces reveal lifetimes as low as several picoseconds, up to 3 orders of magnitude lower than typical lifetimes in bare WSe_2. In addition, photoluminescence maps show quenched emission at the interfaces, whereas photoconductivity is enhanced at the same locations, demonstrating efficient charge transfer from WSe_2 to graphene. From these results, we conclude that the ultrafast quenching of spin/valley lifetime is due to transfer of spin information by the efficiently conducted charge carriers.

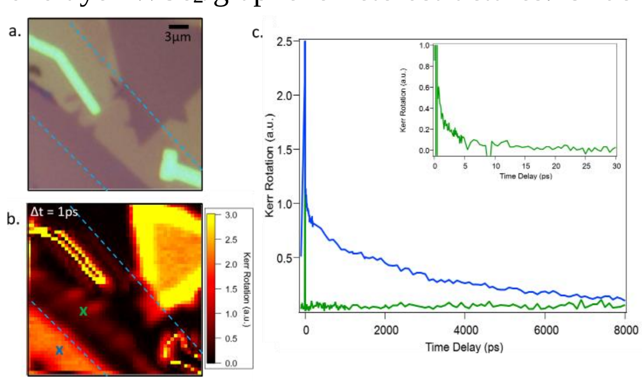

Figure 1. (a) Microscope image of WSe_2/graphene device. Blue lines outline the graphene. (b) TRKR map of spin/valley density at 1ps delay and -30V gate. Kerr signal is suppressed at the heterostructure interface. (c) TRKR delays taken on bare TMD (blue) and interface (green). The inset shows the fast decay at the interface.

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[2] Y. K. Luo, J. Xu, T. Zhu, G. Wu, E. J. McCormick, W. Zhan, M. R. Neupane and R. K. Kawakami. Nano Lett. 17(6), 3877-3883(2017).

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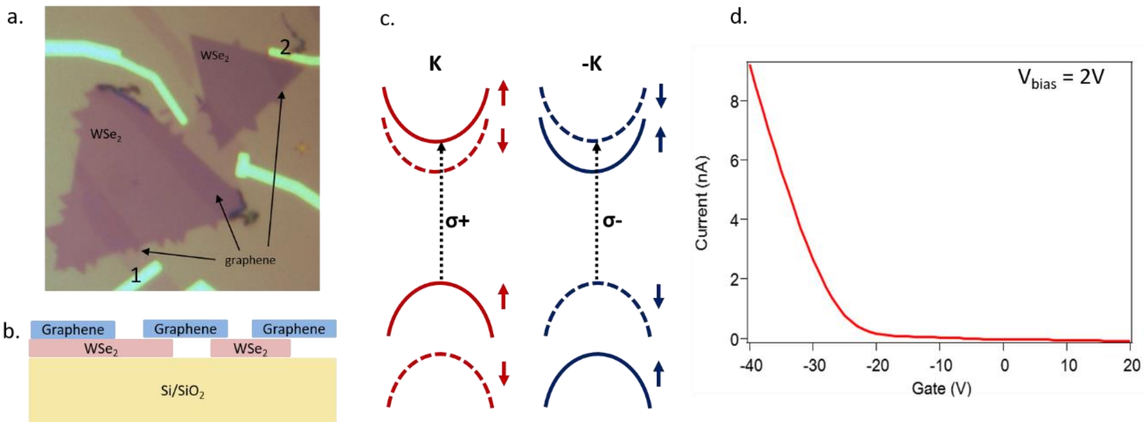
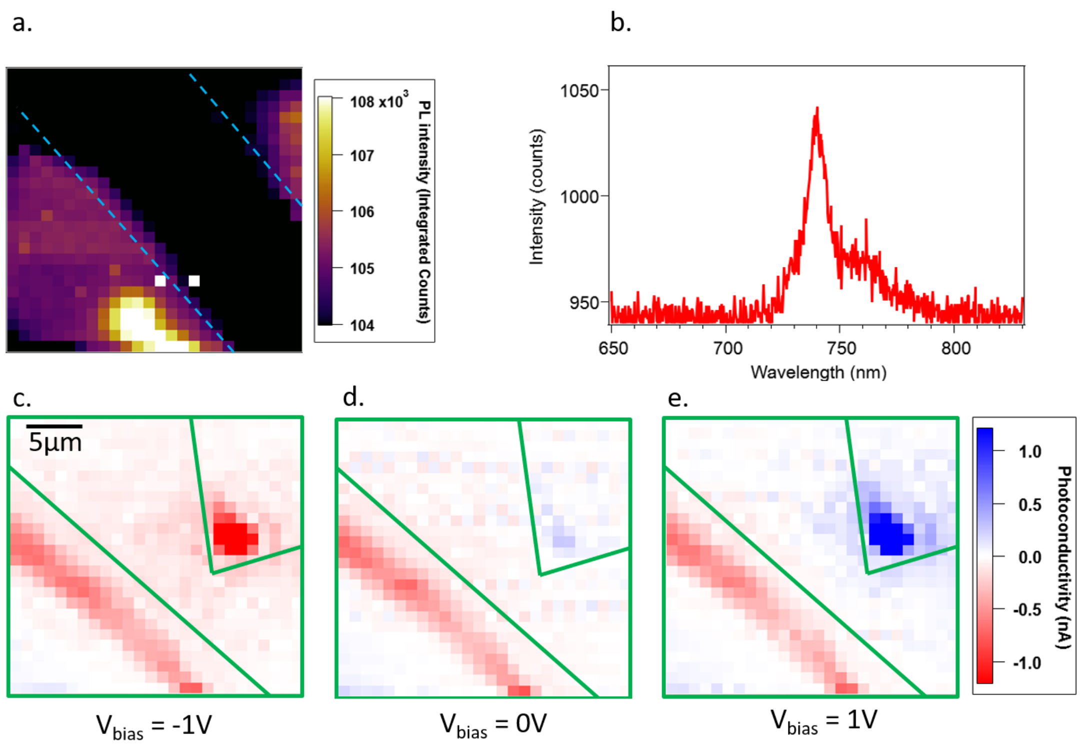
Supplementary Data



Supplementary Fig 1. (a) Optical image of a WSe_2/graphene heterostructure device. The WSe_2 flakes are grown by CVD on SiO_2/Si substrates and graphene flakes are assembled above by polymer-based transfer. (b) Cartoon side view of the device. (c) Band diagram at the K and -K points for WSe_2. Right (left) circular light excites K (-K) valleys respectively. (d) Gate dependent conductivity from contact 1-2 shows that the device turns on at p-type gating.



Supplementary Fig 2. Spatial dependence of photoluminescence (PL) and photoconductivity (PC). (a) Integrated PL map of the WSe_2/graphene heterostructure at -30V gate. Blue lines show approximate graphene edges. Total PL signal is quenched at the interfaces. (b) Typical PL spectrum taken on the bare WSe_2 under excitation by a 532nm laser. (c)-(e) Photoconductivity maps at -1V, 0V, and 1V bias respectively show that PC signal is greatest at the interfaces. At 0V bias the two interfaces contribute in opposite directions, but the application of bias can switch the direction of photocurrent as in (c). We note that the two interfaces do not switch at symmetric bias. All data was taken at -30V gate and 8K. Bias was applied from contact 1-2. Approximate WSe_2 flake outlines are shown in green as a guide to the eye.