Negative Valley Polarization of the Intralayer Exciton via One-Step-Growth of H-type Heterobilayer WS_2/MoS_2

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Abstract

Vertical Type-Ⅱ van der Waals heterobilayers of transition metal dichalcogenides (TMDs) have attracted wide attention due to their distinctive features mostly arising from the emergence of novel electronic structures that include moiré-related phenomena.[1] Owing to strong spin-orbit coupling under noncentrosymmetric environment, TMD heterobilayers host non-equivalent +K and –K valleys of contrasting Berry curvatures, which can be optically controlled by the helicity of optical excitation.[2] The corresponding valley selection rules are well established by not only intralayer excitons but also interlayer excitons.[3] Quite intriguingly, here, we experimentally demonstrate that unusual valley switching can be achieved using the lowest-lying intralayer excitons in H-type heterobilayer WS_2/MoS_2 prepared by one-step growth. This TMD combination provides a very unique case of an ideal interlayer coupling with almost perfect lattice match, thereby also in the momentum space between +K and –K valleys in the H-type heterostructure. The underlying valley-switching mechanism can be understood by bright-to-dark conversion of initially created electrons in the valley of WS_2, followed by interlayer charge transfer to the opposite valley in MoS_2. Our suggested model is also confirmed by the absence of valley switching when the lowest-lying excitons in MoS_2 are directly generated in the heterobilayer. In contrast to the H-type case, we show that no valley switching is observed from R-type heterobilayers prepared by the same method, where interlayer charge transfer does not occur between the opposite valleys. We compare the case with the series of valley polarization data from other heterobilayer combinations obtained under different excitation energies and temperatures. Our unique case of the valley switching mechanism can be utilized for valley manipulation by controlling excitation photon energy together with the photon helicity in valleytronic devices derived from H-type TMD heterobilayers.

References

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