Influence of Defects on the Valley Polarization Properties of Monolayer MoS_2 Grown by Chemical Vapor Deposition

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Abstract
Two-dimensional transition-metal dichalcogenides (TMDs) offers valley degree of freedom, which can be exploited to design next-generation valley-based electronics or "valleytronics". This property enables an exciton to sustain its valley character throughout the time of its existence. In fact, valley polarization approaching 100% has been reported in exfoliated 1L-MoS_2 samples, whereas 1L-MoS_2 films grown by the chemical vapor deposition (CVD) technique, which is frequently used to grow large area films on different substrates, show only moderate polarization values (less than 50%). Since large area coverage of the monolayer film has to be ensured for any practical application of the material, it is imperative to understand the reason for the moderation of valley polarization in CVD-grown 1L-MoS_2. Note that the optical and electrical properties of CVD-grown layers often suffer from the presence of a high density of sulfur vacancy defects (V_S) and the residual strain. Since the valley and spin properties are closely related to the crystal symmetry, both the strain and the defects are expected to have certain impacts on the valley polarization (VP) property of 1L-MoS_2 grown by the CVD technique. Involvement of small wavelength phonons in valley de-polarization of 1L-MoS_2 is an unsettled issue. A recent theory suggests that the long-range part of the electron-hole exchange interaction can virtually transfer excitons between *K* to *K'* valleys without directly involving any phonon [1]. In this process, the momentum scattering of the excitons can influence the spin flip scattering rate through Maialle-Silva-Sham (MSS) mechanism. The presence of defects can influence the momentum relaxation rate of the excitons and hence can affect the valley depolarization. Here, our temperature dependent polarization resolved photoluminescence spectroscopic study experimentally demonstrates, the above mechanism as the most dominant intervalley exciton transfer process in CVD grown monolayers, where momentum scattering of excitons by the air molecules attached to V_S plays significant role [2]. Interestingly, the momentum scattering rate is found to be proportional to the cube root of the defect density. Intervalley scattering of excitons through Γ-valley also contributes to the valley de-polarization process specially when the layer has tensile strain or high density of V_S defects as these perturbations reduces *K* to Γ-energy separation. Band-structural calculations carried out within the density functional theory framework confirm this finding. Experimental results further suggest that exchange interactions with the physisorbed oxygen molecules can also result in the intervalley spin-flip scattering of the excitons and this process becomes important when the defect density is sufficiently high.

References

[1] T. Yu and M. W. Wu, Physical Review B 89 (2014) 205303

[2] F. Mujeeb, et al. Physical Review B 107.11 (2023) 115429