Spin relaxation and intervalley scattering in 2D semiconductors

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Monolayer transition metal dichalcogenides, MX_2 (M = Mo, W and X = S, Se), are direct-gap semiconductors with some interesting properties. First, the low-dimensional hexagonal structure leads to two inequivalent K-points, K and K', in the Brillouin zone. Second, this valley index and spin are intrinsically coupled, and spin-dependent selection rules enable one to independently populate and interrogate a unique K valley with circularly polarized light[1,2].

Here we probe the degree of circular polarization of the emitted photoluminescence as function of the photo-excitation energy and temperature to elucidate spin-dependent inter- and intra-valley relaxation mechanisms. Monolayer flakes of MoS_2 and $MoSe_2$ show a strong depolarization as the excitation energy is increased[3] (Figure). The difference in the excitation energy and photoluminescence emission energy, dE = $E_{pump} - E_{PL}$, governs the depopulation of carriers in each valley. Adding more energy above a distinct threshold characteristic of the longitudinal acoustic (LA) phonon for each material enables inter-valley scattering and produces a sharp decrease in the observed circular polarization. LA phonons in these two systems have different energies (30 meV for MoS_2 and 19 meV for $MoSe_2$), and we show that the threshold for the excess energy required to initiate the depolarization process clearly reflects the material specific phonon energy. In addition, our results show that independent of how many carriers are excited, i.e. whether you create neutral or charged excitons, the scattering process is the same. However, WS_2 maintains significant polarization[4] for high excitation energies, even at room temperature when properly prepared and it will be discussed at the meeting.

References

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