Quadrupolar and dipolar excitons in $MoSe_2$ homo - bilayer

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2D layered semiconductors such as transition metal dichalcogenides emerged as ideal playground to study exciton physics due to greatly enhanced electron hole attraction due to reduced dimensionality and dielectric screening in monolayer limit. Simultaneously, the absence of lattice-matching constraints in van der Waals materials open a new paradigm in material engineering. These materials can be seamlessly stacked into virtually limitless homo- and hetero-structures, whose properties can be finely tuned by material selection and relative orientation. This new type of structures opens the path to explore plethora of exotic physics phenomena including very reach excitonic landscape. For example, homo- and hetero-structures can host dipolar, interlayer, excitons with static dipole moment, which can be than easily tuned by external electric field. Interaction between dipolar excitons can lead to new correlated bosonic states such as Bose – Einstein condensation, exciton superfluidity or generalized Wigner crystal, which can be further tuned by external magnetic field. At the same time, due to the interaction between two dipolar excitons with opposite dipole moments new type of interlayer exciton, namely quadrupolar exciton is expected to form.

Here, we present the first observation of new quasiparticle, the quadrupolar excitons in homo – bilayer of $MoSe_2$. In the reflectance spectra (presented in Fig1 (a)) we observe new features marked by Q1 and Q2 which exhibit quadratic dependence with electrical field (Fig1(b)), a finger print of the quadrupolar excitons due to their composite nature of differently oriented dipole. Electron tunneling as well as dipole dipole interaction, hybridizes the two degenerate, oppositely oriented dipolar excitons into symmetric and antisymmetric quadrupolar excitons, as predicted theoretically (Fig1(c)). Furthermore, theoretical modeling of the excitonic landscape in $MoSe_2$ bilayers reveals not only the formation of quadrupolar excitons and their tunability by electric fields but also their significant impact on the hybridization of intralayer transitions under electric fields, which is in great agreement with our experimental observations. Our result demonstrates that electric field can tune the exciton spieces from intra to interlayer as well as hybrid one.

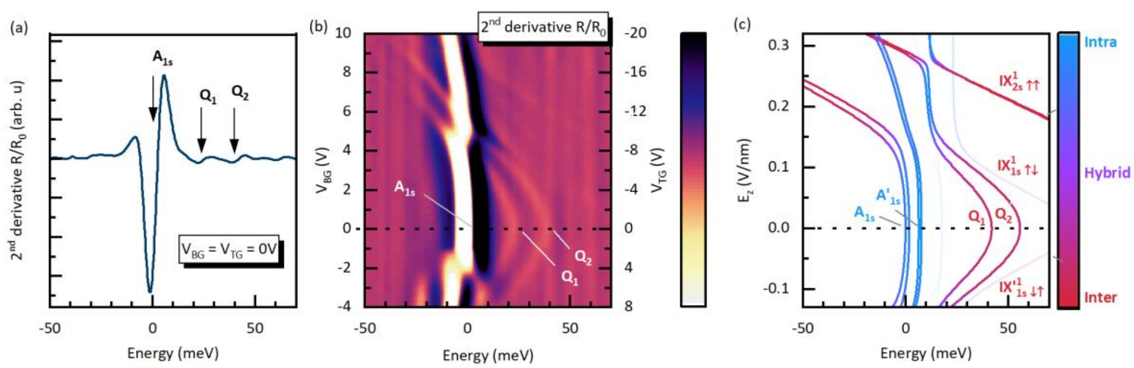

Fig.1 (a) Second derivative of the reflectance spectra, where interlayer and newly form quadrupolar excions (Q1 and Q2) are marked. (b) Second derivative of the reflectance spectra measured as a function of the electric field, where the dipolar character of the Q1 and Q2 is confirmed by characteristic quadratic dependence of the transition energy with electric field. (c) Theoretical calculations of the excitonic landscape as a function of electrical field in bilayer WSe_2, where including coupling mechanism between the interlayer excitons with different dipole moments lead to the formation of quadrupole excitons.