Controlling exciton population in 2D transition metal dichalcogenides

Frank Tseng, Ergun Simesk, and Daniel Gunlycke

Naval Research Laboratory 4555 Overlook St. Washington D.C. 20375 ,USA
The George Washington University, Washington D.C. 20052, USA
frank.tseng.ctr@nrl.navy.mil

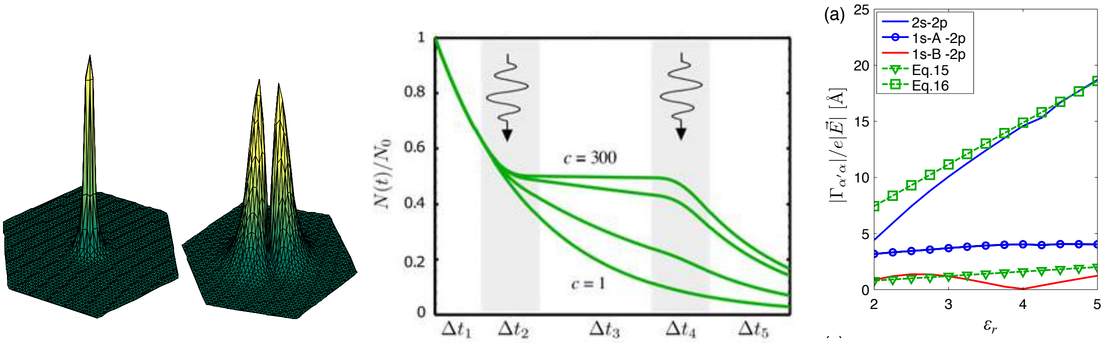
Abstract

Two-dimensional transition metal dichalcogenides (2D-TMDCs) are suitable hosts for excitons [1,2,3], which are an electron/hole pair bound by an attractive Coulomb interaction. The exciton states are both optically bright and dark. Weak dielectric screening and concomitant lattice effects contribute toward large binding energies, and an energy separation between bright and dark states of the same sub-shell. Between the bright 2s and dark 2p state, their energy separation can be greater than a kT of thermal energy at room temperature as it depends on the strength of the Coulomb interaction. Dark exciton states have longer lifetimes inherent to their symmetry which spatially separates the electron and hole (cf. Fig. 1a), and inhibits radiative recombination. Therefore, an optically accessible dark 2p state in these 2D materials is useful for energy management applications and could potentially enable Bose-Einstein condensates, for excitons.

We present a model for controlling an exciton population mediated through a longer-lived dark 2p state, as shown in Fig.1b. To characterize these transitions, we calculate the needed transition matrix element based on exciton states determined using a minimalistic Triangular Lattice Exciton (3-ALE) model [2]. For 2D-TMDCs, the closest bright states to the dark 2p state are 1s-A, 1s-B and 2s. The split 1s ground states arises from a combination of spin-orbit coupling, onsite Coulomb interaction and relative permittivity [3]. We find the transition matrix element describing transitions from the ground states to weakly depend on the dielectric constant. Meanwhile the matrix element between 2p and 2s is strongly dependent on the dielectric constant as shown in Fig.1c. The strength of the matrix element is an important consideration in controlling the exciton population to and from the dark 2p state, as it is inversely related to the Rabi-oscillation period and the time-scale for complete population transitions between states.

References

[1] R.F. Frindt and A.D. Yoffe, Proc. R. Soc. Lon. Ser-A., **273** (1963) 69-83.
[2] F.C. Wu, F.Y. Qu and A.H. MacDonald, Phys. Rev. B. **91** (2015) 075310.
[3] D. Gunlycke and F. Tseng, arXiv:1504.04040 (2015).



(a) **(b)** **(c)**

Figures

Figure 1. (a) Real-space probability density of an electron relative to the hole at the center of the lattice for 1s-A and 2p exciton states. (b) Schematic of the lowest exciton states and calculated time-dependent progression of exciton population to and from a dark p state for a range of potential lifetimes. (c) Transition matrix elements as a function of relative permittivity for MoS_2. Corresponding 2D-hydrogen model calculations in dashed-lines.