Selective photo-excitation and angular momentum imprint of exciton complexes in 2D materials by using twisted lights

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In this work, we will review our recent experiment-theory-joint studies of the light-matter interactions between excitons in atomically thin transition-metal dichalcogenide (TMD) monolayers and spatially structured lights, including twisted lights (TLs) carrying orbital angular momenta (OAM) and spin-orbit-coupled vector vortex beams (VVB).

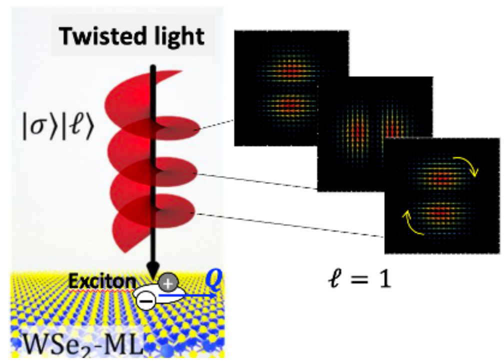

Fig. 1. Schematics of a twisted light that carries an orbital angular momentum, $\ell\hbar$, normally incident to a 2D WSe_2 monolayer and photo-generates an exciton with the center-of-mass momentum, $\boldsymbol{Q}$. The square insets show the spatially structured patterns of the vector potential of the incident TL in the different cross-section planes.

Twisted light is a type of specially structued light where the wavefront is spatially twisted so as to acquire OAM, being a new degree of freedom of light in addition to that of spin angular momentum (SAM), i.e. polarization. Note that the quantum number of the optical OAM carried by a TL, unlike SAM, can be an arbitray integer and is prospective for the realizing the high-capacity quantum information technology and high dimensional quantum entangled states.

A few years ago, we for the first time observed the photoluminescences (PLs) from a hBN-encapsulated MoS_2-ML under the excitation of TL with controlled OAM, which are shown blue-shifted with the increment of OAM of the incident TL.[1] Theoretically, we developed the theory of light-matter interaction between 2D excitons and TLs within the framework of time-dependent perturbation theory. The band structures and transition dipoles of 2D excitons, including bright excitons and various dark excitons, in MoS_2 and WSe_2 monolayers are numerically calculated by solving the first-principles-based Bethe Salpeter equation (BSE). [2] By using the developed theory, the observed blue-shifts of PLs are realized as a consequence of selective photoexcitation of finite momentum exciton states by the incident TLs with OAMs. Those TL-generated exciton states are simulated and shown to form localized exciton wave packets encoded by OAM, suggested to be deciphered by using angle-resolved optical spectroscopy. [3]

Our developed theory of TL-exciton interaction is further extended and applied to the studies of vector vortex beam (VVB) formed by the superposition of two distinct TLs with opposite SAM and OAM. Since the SAM and OAM in a structured light are inherently coupled by the optical spin-orbit interaction (SOI), the vectorial polarization of a VVB is no longer purely transverse but possesses the longitudinal component. The existence of a longitudinal field in a spin-orbit-coupled VVB enables the optically activation of one of the dark exciton doublets of TMD-MLs, namely the gray exciton. We show that the photo-excitation of gray excitons can be enhanced by increasing the OAM of VVB and is robust in the OAM transfer from a VVB against decoherence.

References

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