Altering the Dimensionality of Exciton-Exciton Annihilation in atomically thin Black Phosphorus

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Dimensionality plays an important role in determining the fundamental nature of excitons as well as their interactions. For example, the binding energy of excitons is orders of magnitude higher in 1D (carbon nanotube-CNT) or 2D (transition metal dichalcogenides-TMDC) semiconductors as compared to conventional 3D semiconductor (Gallium Arsenide-GaAs) [1,2]. Not only that, signature of excitonic interaction is strikingly different for 1D systems as compared to 2D/3D systems [3]. In 1D systems (CNTs) the rate of exciton-exciton annihilation deviates from the mean filed approximation as the excitons are constrained by the physical dimensions to interact only with their nearest neighbors resulting in a time dependent rate [4]. However, in 2D (TMDC) and 3D (GaAs), the dimensional restrictions are less stringent resulting in a time independent constant rate of annihilation consistent with the mean field approximation [5]. A particularly interesting case is presented by excitons in black phosphorus (BP), where quasi 1D excitons have been observed in atomically thin sheets of BP, resulting in a unique system of 1D excitons in a 2D plane [6,7]. Here we show that the interaction between excitons in atomically thin BP shows both 1D as well as 2D characteristics depending on the experimental parameters such as exciton density and temperature. We use µ- transient absorption spectroscopy(µ-TAS) to study the exciton-exciton annihilation process in bilayer (2L) BP. We observe the classic 1D time dependent exciton-exciton annihilation dynamics at low exciton density and with increasing exciton density the data shows time independent rate, characteristic of a 2D exciton-exciton annihilation. We also observe more 1D characteristics at low temperature for all exciton densities. We attribute this effect to the anisotropic diffusion of excitons in atomically thin BP. Our data matches well with the phenomenological model of anisotropic diffusion limited exciton-exciton annihilation [8]. Our work highlights the importance of atomically thin black phosphorus as a unique platform to study various interesting many body excitonic interactions.

References:

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