Signature of universal grammars in solvated 2D materials

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Dimension reduction of materials is the holy grail that leads to abnormal or anomalous physics of systems. Especially, fruitful electronic pictures are attractive to physicists and chemists owing to an unprecedented potential that was not expected in 3D or quasi-2D phases. Since nanotechnology appeared, it has been disclosed that 2D materials beyond graphene show distinct optical or electronic behaviors. However, there is rarely work to shed light on the physics when they are solvated in solutions. This can be originated from interfacial phenomena. In detail, the solvent effect which is mystified still should be described carefully while the 2D materials need to be treated by post- or beyond-DFT theory since excitonic behavior is not ignorable in this dimension.

To resolve this issue, we developed a new theory where GW approximation and generalized Poisson equation are combined without any empirical parameters, abbreviated GW-GPE.1 In this framework, the solvent effect is decoupled into polarization-field effect (PFE) and environmental screening effect (ESE). By using this method, we first established the structure-property relationship. Surprisingly, the hole sizes in percolated structures are strongly correlated with both effects. Finally, we aligned band edge positions of 2D materials and compared these with experimental observations or measurements, showing excellent agreement.

Next, we extended our theory to solve excitonic problems by conjugating Bethe-Salpeter equation (BSE) to GW-GPE, that is, GW/BSE-GPE. With this theory, we discovered the universality of 2D materials in solutions. First, band gap changes by PFE are exponentially scaled with dielectric constants of solvents regardless of materials and scales of band gap changes. Second, the optical band gaps, that is, the first bright peaks in the absorption spectra are stationary as the dielectric constants vary. In other words, a solvatochromic picture can be broken out in low-dimensionality. These two findings suggest that there will be physics that worked universally in solvated 2D materials while implying this new theory can be an unveiler for hidden physics of low-dimensional materials in solutions.

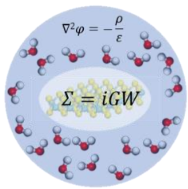

Figure 1 Schematic of GW-GPE

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References

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