Testing the HSE functional for defects in layered van der Waals materials.

Hexagonal boron nitride, h-BN, as a layered van der Waals material with a structure similar to graphite or, in monolayer form, similar to graphene, has attracted considerable attention in recent years. Due to its wide band gap, h-BN is a potential candidate for optoelectronic applications in the ultraviolet energy range[1-2]. The study of defects in h-BN, has demonstrated polarized and ultra-bright single-photon emission in two-dimensional h-BN (2D h-BN)[3] at room temperature, having the potentially for better scalability than the long-studied nitrogen-vacancy center in diamond. It indicates the possibility of applying two-dimensional materials in quantum information technology and nanophotonics.

On the theoretical side, the standard implementations of density functional theory, local-density- and generalized-gradient-approximation (LDA and GGA, respectively), fail to describe defects in wide band-gap materials due to an artificial delocalization of defect states. This has its origin in their failure to produce the physically correct linear behavior of the total energy with respect to the fractional occupation numbers[4-5]. (In other words: the generalized Koopmans' theorem[4] is not satisfied.). In a recent study on Ga_2O_3[6], it has been shown that the parameters of a Heyd-Scuseria-Ernzerhof (HSE)[7] hybrid functional can be optimized to overcome this issue, and to reproduce the band gap at the same time. We provide a test of this procedure for bulk h-BN and 2D h-BN, which has strong anisotropic screening.

Our results show that the use of a hybrid functional with tuned coefficients is successful in fulfilling the generalized Koopmans theorem for h-BN in bulk, and the combination of α=0.4 and μ= 0.3 results in a band gap of 5.99 eV in agreement with GW calculations and experiment[8,9]. We have checked these parameters in 2D h-BN (using the SLABCC charge correction code[10], the implementation of the Komsa-Pasquarello method for total energy correction for charged periodic slab models)and have found that the band gap and the linearity criterion are not satisfied. We have not been able to find any other set of parameters two fulfill both criteria in 2D h-BN, probably due to the very anisotropic screening.

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