GW(Γ) calculation for photoabsorption energies of spin polarized systems

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There has been an increasing demand of using the GW method[1] based on many-body perturbation theory not only in the field of solid-state physics but also in the field of quantum chemistry, and many codes installing this method have been developed recently. However, the usual GW method enables one to calculate photoemission and inverse photoemission spectra (PES/IPES) only. To calculate photoabsorption energies (PAEs), one had to additionally solve the Bethe–Salpeter equation (BSE). Since this method combines the GW calculation and the BSE calculation, this method is called GW +BSE method[2]. This method should deal with not only the one-particle Green's function in the GW part but also the two-particle (electron-hole) Green's function in the BSE part, and doubly induces the evaluation errors. It has been recently reported that the GW + BSE method significantly underestimates the experimental PAEs of small molecules[3]. To avoid these problems, it is highly desirable to develop a new GW method to calculate PAE without solving the BSE. The idea is very simple: Just to use the pure GW method to the cationic systems not to the neutral systems. Then, one may obtain the PAE from the quasiparticle (QP) energy differences between the (one electron missing) hole level and the higher empty levels. It enables one to identify the rigorous one-to-one correspondence between the PAE peak and the ("extended") QP level. This simple idea yields the PAE with a reasonable accuracy. This method is particularly useful for radical systems but is applicable for both closed shell and open shell atoms and molecules. We applied the self-consistent LGWΓ[4] method including the vertex correction to our method. Figure 1 shows the absolute difference between the calculated and experimental values of the first photoabsorption energy. As seen in Figure 1, this method gives PAEs within 0.1 eV accuracy.

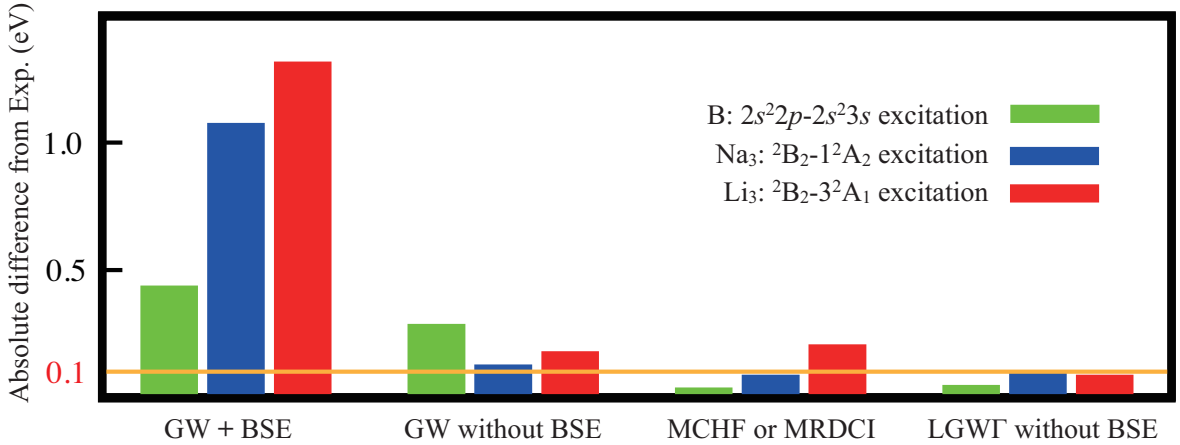

Figure 1: First photoabsorption energies of B, Na_3, and Li_3[5]

References

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