Calculation of Positron Binding Energies Using the Generalized any Particle Propagator Theory

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We recently extended the electron propagator theory to any type of quantum species based in the framework of the Any-Particle Molecular Orbital (APMO) approach [1-3]. This generalized APMO propagator theory (APMO/PT) was applied to calculate positron binding energies (PBEs) of anions and some diatomic molecules using the second order, third order, and renormalized third order quasiparticle APMO/PT approaches and compared our results with those previously calculated employing configuration interaction (CI), explicitly correlated and quantum Montecarlo methodologies. We found that renormalized APMO/PT methods can achieve accuracies of ⏃0.35 eV for anionic systems, compared to Full-CI results, and provide a quantitative description of positron binding to anionic and highly polar species.

We have employed thermodynamic cycles to relate calculated APMO/PT PBEs with the stability of homo- and heteronuclear $e^+[X^-Y^-]$ compounds formed by two halide anions ($X^-,Y^- = F^-,Cl^-,Br^-$) and one positron. Our results indicate the formation of energetically stable positron molecules in which the formation of positron covalent bonds underlie the stabilization of the otherwise repelling dihalides, similar to the bond observed in the $e^+[H_2^{2-}]$ complex [4]. Surprisingly, several aspects of the positronic dihalides are similar to those of the purely electronic analogs, $e^-[A^+B^+]$, molecular cations with isoelectronic atomic cores ($A^+,B^+ = Na^+$, K^+, Rb^+) bound by one electron.

Third order APMO/PT approaches display a considerable potential for studying positron binding and positron bonds in large molecules as they provide a reasonable compromise between computational effort and accuracy in view of the well-known difficulties to account for electron-positron correlation.

References
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