Many-body theory of positron interactions with polyatomic molecules

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Positrons are unique probes of matter, with applications in materials science (ultra-sensitive diagnostic studies of surfaces, defects and porosity), medical imaging (PET), astrophysics, molecular spectroscopy [1], and fundamental physics. Positron interactions with matter are characterised by strong many-body correlations. They significantly modify scattering, and enhance annihilation rates by orders of magnitudes (see [2] for a review). They also make the theoretical study of positron-matter interactions very challenging.

We have developed a diagrammatic many-body description of positron-molecule interactions that takes ab initio account of the correlations, implemented in the state-of-the-art code `EXCITON+` [3]. We solve the Dyson equation for the positron quasiparticle wavefunction in a Gaussian basis, constructing the positron-molecule self-energy including the *GW* diagram (at RPA/TDHF/BSE levels of theory), describing polarisation, screening and electron-hole attraction interactions, the ladder series of positron-electron interactions that describes virtual positronium formation, and the ladder series of positron-hole interactions. We have used it to calculate binding energies for a range of polar and non-polar molecules, focusing chiefly on the molecules for which both theory and experiment exist [3,4], but also making predictions (e.g. of positron binding to DNA nucleobases [3], and of the effect of fluorination vs chlorination in hydrocarbons [4]). Delineating the effects of the correlations, we show, in particular, that virtual-positronium formation significantly enhances binding in polar molecules, and moreover, that it can be essential to support binding in non-polar molecules. Overall, we find the best agreement with experiment to date (to within a few percent in cases). We have recently developed and extended the method to the calculation of the positron scattering [5] and annihilation gamma spectra in molecules [6] providing insight that should support the development of fundamental experiments and the myriad of antimatter-based technologies and applications. Moreover, the positron-molecule problem provides a testbed for the development of methods to tackle the quantum many-body problem, for which our results can serve as benchmarks.

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