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Positron binding and annihilation in molecules

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State of the art and motivations

The positron is the antiparticle of the electron. It was the first *antimatter* particle ever discovered, first theoretically by Paul Dirac in 1931, and then experimentally by Carl Anderson in 1932, both physicists awarded the Nobel Prize soon after. Positrons are also the simplest and most abundant form of antimatter. They come from β^+ radioactive decays, can be generated in accelerators, and are produced in large quantities (15×10^9 tonnes per second!) near the centre of our Galaxy. The ability of positrons to *annihilate* with electrons and emit characteristic annihilation gamma rays, underpins their use in various diagnostics, from positron lifetime spectroscopy of solids to positron emission tomography (PET) in medicine. When partnered with antiprotons, positrons can form *antihydrogen*, currently under intense investigation at CERN.

The electrons with which positrons annihilate are usually not free but packed in atoms or molecules. As a result, the process of positron annihilation is strongly affected by the positron interaction with the target. For example, positrons are repelled by atomic nuclei, so they usually annihilate only with the outermost, valence electrons. On the other hand, when a positron approaches an atom or molecule, it *polarises* (i.e., distorts) the electron cloud. This gives rise to an attractive polarisation potential acting on the positron. Another important effect is "hopping" of an atomic electron to the positron, temporarily forming an electron-positron "atom" called positronium (Ps). This increases the positron-atom attraction and strongly enhances the positron annihilation probability (see, e.g., detailed calculations for noble-gas atoms [1]).

For many atoms, the attraction is so strong that it overcomes the positron-nucleus repulsion and allows the creation of positron-atom *bound states* (predicted in [2] and proved variationally in [3]). To date, positron binding to about ten atoms has been predicted in state-of-the-art calculations. There are also firm expectations that many more atoms are capable of binding [4, 5], but there have not been any experimental conformation of this phenomenon yet [6].

What makes the problem of binding so important is the effect it has on positron annihilation in molecules [7]. When a positron collides with a molecule, it can be captured into the bound state by transferring its excess energy into molecular vibrations. This gives rise to *resonances* and orders-of-magnitude enhancement of the annihilation rates. Annihilation studies of resonances have enabled measurements of positron binding energies for more than 80 molecules (see [8] and references therein). In contrast, a significant theoretical effort towards computing positron-molecule bound states resulted in only a handful of predictions that can be compared with experiment, with the best agreement being at 25% level [9]. Also, most of the calculations performed to date considered strongly polar molecules (i.e., those with a large permanent dipole moment), because positron binding to these species is easier to describe. Model calculations [10] aside, there is very little theoretical understanding of positron binding to nonpolar molecules.

Objectives & Methodology

The aim of the project is to explore positron binding to molecules by developing a new approach to this difficult problem. The positron-molecule attraction and binding are due to electron-positron *correlation effects*, such as polarisation and virtual Ps formation. These effects are notoriously difficult to describe theoretically. Even modern quantum chemistry approaches cannot provide the accuracy required for the calculation of small positron-molecule binding energies (from few to a few tens of millielectronvolts), making reliable *ab initio* calculations impossible. On the other hand, the main features of the positron-molecule interaction are clear, e.g., from our extensive studies of positron-atom interactions [1]. Based on this, one can construct physically meaningful positron-molecule correlation potentials that will contain one or two adjustable parameters [11]. These parameters are chosen by comparison with existing high-quality calculations or by using experimental data for a small subset of molecules. The correlation potentials thus determined, positron bound state energies and wavefunctions will then be computed for a wide range of molecules. This should allow us to obtain theoretical understanding of many trends of positron-molecule binding, such as linear scaling with molecular dipole polarisability and dipole moment (surpassing the results of simple modelling of the latter [12]); see our latest work [13].

On the technical side, calculations of positron binding and annihilation rates are done by expanding the capabilities of GAMESS [14], an advanced, free quantum-chemistry package. This work is currently led by Dr Andrew Swann, a post-doctoral research associate, supported by the EPSRC grant "Positron bound states and annihilation in polyatomic molecules", with Dr Gribakin being the principal investigator. A very interesting extension of the work will be computation of positron-molecule annihilation gamma-ray spectra, an area where large amounts of experimental data await proper theoretical understanding.

Collaborations

Our group has a successful long-term collaboration with the experimentalist group of Prof. Cliff Surko (University of California, San Diego), whose famous positron trap is the workhorse of many positron experiments world over, and who pioneered measurements of positron-molecule resonant annihilation and binding energies. New links are emerging with the group of Dr. David Cassidy at UCL (London), who develop novel experiments with long-lived Rydberg-state positronium, with prospects of measuring positron binding to atoms and molecules [6]. There are also prospects of further collaboration with Professor John Stanton on the role of anharmonic effects and multimode vibrations in positron annihilation [15].

Required skills

The candidate is expected to have good working knowledge of and interest in Quantum Mechanics. He/she will be expected to learn and master quantum-chemistry computational approaches. Running, modifying and developing computer codes will be an integral and substantial part of the project. In spite of the complexity of the codes, the underlying physics of positron-molecule binding and positron annihilation is relatively simple, and it is expected that many interesting insights into the problem will be obtained.

Further information

While focussed on the positron-molecule binding problem, the project will inroduce the candidate to the range of problems concerning antimatter and its applications, as well as to methods of quantum chemistry and its computational approaches. The experience of handling and writing computer codes, that will be acquired through the work on the project, will be useful in a wide range of future careers.

For further information, please contact Dr Gleb Gribakin.

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