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# Many-body theory of positron interactions with molecules/condensed matter

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*State of the art and motivations*

Positrons are the *anti-particles* of electrons, having the same mass, but opposite, positive charge. They are the simplest form of antimatter. Positrons are produced in abundance in our Galaxy, and are readily obtained in laboratories using accelerators or radioactive isotopes. When positrons come into contact with their matter counterpart, the pair can *annihilate* in a pyrotechnic flash, releasing their energy as a burst of light in the form of two characteristic detectable gamma-rays.

As such, they have important use in medical imaging in PET (Positron Emission Tomography) scans, for diagnostics of industrially important materials, and in understanding antimatter in the Universe. Fundamental interactions of positrons and more complicated antimatter (e.g., antihydrogen) with atomic systems are under intense investigation in numerous international laboratories including CERN, to illuminate collision phenomena and perform precision tests of the standard model and gravity.

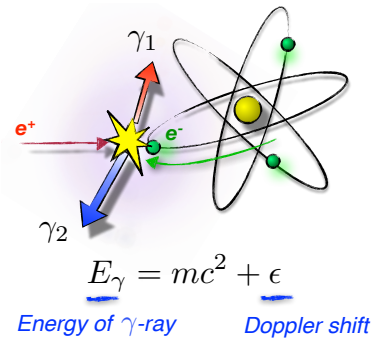


**Fig. 1:** Positrons annihilate with electrons in atoms producing two detectable $\gamma$-rays whose energies are Doppler shifted by an amount characteristic of the electron state involved. Positron annihilation is a unique probe for material characterisation, medical imaging and astrophysics.

When low-energy positrons interact with normal matter, such as atoms, they pull strongly on the electrons and may even cause one of the electrons to 'dance' around the positron, forming so-called positronium (as the positron and electron may annihilate, this may ultimately be a 'dance to the death'). Such effects are known collectively as *correlations*. Correlations have a very strong effect on positron interactions with atoms and molecules. In particular, they can enhance the rate of positron annihilation by many orders of magnitude. They also make accurate theoretical description of the positron-atom system a *challenging many-body problem*.

Proper interpretation and development of the difficult fundamental experiments, materials science techniques and wider applications (e.g., PET) relies heavily on calculations that accurately account for the correlations. Calculations of positron scattering, cooling and annihilation in atomic and molecular gases are crucial for the development of efficient traps and accumulators, used in e.g., the antihydrogen experiments at CERN, and for ultra-high-energy-resolution positron beams that can probe resonant annihilation phenomena and provide a unique tool

for molecular spectroscopy. They can be used to determine electron momentum densities from positron annihilation measurements, may help explain the composition of the interstellar medium by studying the strong $\gamma$-ray signal from the galactic centre, and develop PET, e.g., by improving understanding of energy deposition and dosimetry, or via development of a new spectroscopic PET.

A powerful method of describing positron interactions with many-electron atoms and molecules, which allows for the study and inclusion of interactions in a natural, transparent and systematic way, is *diagrammatic many-body theory* (see, e.g., [1–3]). In this method, instead of computing the complicated many-particle wavefunction, processes of interest — e.g., positron annihilation with an atomic electron — are represented by a series of relatively simple and intuitive diagrams that describe the most physically important contributions to the quantum amplitude of the process. Diagrams are constructed systematically according to a strict set of rules governed by the fundamental nature of the interactions. By applying a 'dictionary' to the resulting diagrams, corresponding analytic expressions can be readily generated and their contribution to the amplitude calculated numerically.

We have had considerable success developing and applying many-body theory of positron interactions with atoms, most notably providing a complete description of the positron interactions with noble-gas atoms [2–6], and very recently the first accurate *ab initio* calculations for positronium (an atom consisting of a bound electron-positron pair) interactions with atoms [7]. Significant development is, however, required to enable accurate calculations for positron interaction with molecules and condensed matter. Sophisticated methods such as R-matrix fail to accurately describe positron annihilation rates even for the simplest molecule, $H_2$ (see e.g., Table 2 of [8]). For condensed matter, although much understanding has been gained by applying *density functional theory*, it fails to reproduce the measured properties of positron interactions with simple systems, such as the noble-gas solids [9].

*Objectives & Methodology*

This aim of this project is to incorporate positrons into existing state-of-the-art many-body theory based electronic structure codes [10, 11][1] to enable the accurate *ab initio* description of positron interactions (binding, scattering and annihilation) with either (1) molecules, or (2) condensed matter, the choice depending on the student's experience and interests.

(1) **Many-body theory of positron interactions with molecules** (*lead supervisor Dr Green, 2nd supervisor Dr Gribakin*). Calculations will first be performed in the mean field (Hartree-Fock) approximation, with the results compared against existing calculations performed by our group using the GAMESS quantum-chemistry code [12][2]. Following this benchmarking, we will develop and employ the full many-body theory approach for *ab initio* calculations, taking account of the correlations including polarisation of the molecular electron cloud and virtual-positronium formation. We will consider small molecules (e.g., $H_2$, $N_2$, $CH_4$), for which we can compare our results to both our previous model calculations that crudely attempted to account for correlations [8], and to experiment (long-standing measurements of $\gamma$ spectra for over 50 molecules [13] still remain totally unsupported by theory). Focussing on small molecules will provide a lot to do and explore, and will elucidate the important fundamental interactions. The successful delivery of the project will pave the way for future calculations on larger molecules (e.g., polyaromatic hydrocarbons, which

[1] These codes were initially developed to study *purely electronic* properties and processes in molecules and condensed matter.

[2] Our group has already incorporated positrons into the GAMESS package. However, GAMESS does not currently include capability for many-body theory calculations, in contrast to EXCITON or YAMBO, which are specifically many-body theory based codes.

contribute to the $\gamma$-ray signal from the galactic centre; and DNA base pairs, important for understanding positron interactions in biological matter and to develop PET technologies).

(2) **Many-body theory of positron interactions with condensed matter** (*lead supervisor Dr Green, 2nd supervisor Dr Grüning*). *If the candidate has appropriate experience in theoretical or computational condensed matter physics*, then they may alternatively develop the many-body theory for positron interactions with condensed matter. In this project we will initially focus on the noble-gas solids, for which there is currently a huge discord between the calculated values (density functional theory and model potential calculations) vs. experimental data of the positron energy levels (workfunctions and affinities) (see Table III of [9]). In the first approach, we will model the system using the '*atomic superposition potential*' approach, solving the Schrodinger equation for a positron in the potential $V_+(\mathbf{r}) = \sum_{\mathbf{R}} V^{(0)}_{\rm atom}(|\mathbf{r}-\mathbf{R}|) + V_{\rm corr}(\mathbf{r})$, where $\mathbf{r}$ is the positron coordinate, the $\mathbf{R}$ summations run over the positions of the nuclei, $V^{(0)}_{\rm atom}$ is the Coulomb (or Hartree-Fock) potential of the positron in the field of the atom, and $V_{\rm corr}$ is the correlation potential known from our positron-atom work. Calculations of the band structure, positron affinities, and annihilation momentum densities and rates will be performed. We will then develop the codes to enable *ab initio* calculation of these quantities using many-body theory.

*Collaborations*

Our group has internationally leading expertise in positron and many-body theory, and we have strong links to the leading experimental groups, e.g., the pioneering University of California San Diego group of Prof. Cliff Surko. Direct collaboration is envisaged with experts in computational many-body physics, including the electronic structure group at Trinity College Dublin.

*Required skills*

The candidate is expected to have good working knowledge of and interest in Quantum Mechanics. To undertake the project focussed on molecules this is the only pre-requisite. For the project focussed on condensed matter, the candidate should additionally have experience and interest in condensed matter theory. In both cases he/she will be expected to learn and master many-body theory methods and their application to many-electron systems. Running, modifying and developing sophisticated computer codes is an integral and substantial part of the project. In spite of the complexity of the codes, the underlying many-body theory should allow great transparency and good physical understanding of the results.

*Further information*

The student will contribute to a vibrant team that is expected to consist of at least two postdoctoral researchers and multiple PhD students delivering the objectives of the European Research Council project ANTI-ATOM: "*Many-body theory of antimatter interactions with atoms, molecules and condensed matter*" led by Dr Green. At least one postdoctoral researcher will be working on the topic, with whom the student can closely interact.

The student will develop a diverse set of skills and acquire broad experience, including in atomic and molecular scattering theory, positron physics, and many-body theory. The latter possess great universality and are used (in slightly different form) in areas ranging from elementary particle theory, nuclear theory and condensed matter theory, thus increasing options for postdoctoral research. The experience of handling and writing computer codes, which will be acquired through the work on the project, will be useful in a wide range of future careers.

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