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The POCOBIO Database for Computed Scattering Cross-Sections for Positron Collisions with Biomolecular Systems

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The design of a database for positron interactions with biomolecular systems is outlined. The database contains only scattering cross-sections, which are derived from theory. The data model is defined in a very flexible way, which facilitates the usage of weakly bound clusters of molecules and molecular systems with many tautomeric forms.

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1. Introduction

In recent years a growing number of databases has been developed to provide repositories for scattering cross sections and reaction rates for collision processes of electrons and heavy particles with atoms and molecules. A recent example with a focus on plasma processing is the open-access website LXcat (pronounced "elecscat") by Pitchford et al. [1] within the Plasma Data Exchange Project. LXcat is a collection of currently 22 databases. Another recent exmample is the Quantemol database (QDB) by Tennyson et al. [2]. Two databases with a focus on astrochemistry are the Phys4Entry database by Celiberto et al. [3] and the KInetic Database for Astrochemistry (KIDA) by Wakelam et al. [4].

Several databases with a focus on interactions of ions, electrons, positrons and photons with biomolecular systems are organised in the RADAM (RADiation DAMage) database portal (see e.g. Denifl et al. [5]). The RADAM roadmap [5] is suggesting the creation of a database for positron interactions with biomolecular systems. However, collections of data for collision processes of positrons with atoms and molecules are mainly presented in review articles, but not in databases, see e.g. Surko et al. [6], Danielson et al. [7] and Brunger et al. [8].

The purpose of this paper is to present the POCOBIO database for positron collisions with biomolecular systems. The POCOBIO database will only contain theoretical cross section data from calculations. It will not contain any experimental data This allows to have a consistent set of data. The database should fulfill several purposes:

- work as a repository for cross section data,
- allow easy access so that the data can be used in computer program packages for simulation of positron tracks,
- provide benchmark data for other calculations and for experimentalists,
- and provide differential cross sections, which allow experimentalists to correct experimentally obtained integral cross sections.

Differential cross section data can be included for a fine grained angular grid, which is not possible for tabular data in usual publications. If differential cross sections are available from theory, these can be used to calculate angular corrections to experimental data for gas-phase experiments. Most of the experimentally available integral cross sections are obtained with the linear transmission technique. In such experiments it is not possible to distiguish between unscattered particles and particles that are scattered within a certain angular cone in forward direction (and in some cases also in backward direction). This phenomenon is also called angular discrimination error. The ifferential cross section data in the POCOBIO database is aimed to simplify the angular correction in experimental integral cross sections.

The paper is organized as follows. Section 2 will give a short overview of the physical processes. Section 3 lists the current data sources. In Section 4 the data model is discussed. Section 5 gives an outlook on future developments of the database.

2. Overview

Some of the most important processes between positrons and molecules are given below. In the following M is a biomolecular system. Here a biomolecular systems is understood to be either a single molecule or a cluster of several weakly bound molecules. The labels (n_i, ν_i, j_i) indicate the initial electronic, vibrational and rotational state. Similarly the labels (n_f, ν_f, j_f) describe the final

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state for the neutral molecule and the labels (n_f^+, ν_f^+, j_f^+) are for the final state of the molecular ion M^+. The state of Positronium is given by the label n_{Ps}.

- elastic scattering:
$$M(n_i, \nu_i, j_i) + \mathrm{e}^+ \rightarrow M(n_i, \nu_i, j_i) + \mathrm{e}^+$$
- inelastic scattering:
$$M(n_i, \nu_i, j_i) + \mathrm{e}^+ \rightarrow M(n_f, \nu_f, j_f) + \mathrm{e}^+$$
- ionization:
$$M(n_i, \nu_i, j_i) + \mathrm{e}^+ \rightarrow M^+(n_f^+, \nu_f^+, j_f^+) + \mathrm{e}^+ + \mathrm{e}^-$$
- annihilation:
$$M(n_i, \nu_i, j_i) + \mathrm{e}^+ \rightarrow M^+(n_f^+, \nu_f^+, j_f^+) + 2\gamma + 3\gamma + \cdots$$
- Positronium formation:
$$M(n_i, \nu_i, j_i) + \mathrm{e}^+ \rightarrow M^+(n_f^+, \nu_f^+, j_f^+) + Ps(n_{\mathrm{Ps}})$$

Here we do not consider dissociation of the molecular system, because with the currently available theoretical tools and computer software it is not easy to calculate these processes. In general we will assume that the neutral system M and the ionic system M^+ can be described by the same rotational and vibrational modes.

3. Data sources

Currently we have data available for the positron collisions with the following biomolecules: pyrimidine [9], tetrahydrofuran (THF) [10], uracil [11] and the DNA nucleobases adenine, guanine, thymine and cytosine [12]. The last manuscript includes cross section data for 6 tautomers of adenine, for 9 tautomers of guanine and for 5 tautomers of cytosine. THF is included here, because it is considered as a model compound for the sugar in the backbone of the DNA (see e.g. Boudaiffa et al. [13] and Sanche [14]). For pyrimidine and THF experimental scattering cross sections are available. Cross sections for positron collisions with pyrimidine in the gas phase have been measured by Zecca et al. [15] in Trento and by Palihawadana et al. [16] at the Australian National University (ANU). Cross sections for positron collisions with THF in the gas phase have been measured by Zecca et al. [17] in Trento and by Chiari et al. [18] at ANU. For both molecules the computed differential cross sections compare well with the experimental data. The errors are usually below 10 per cent. In Franz and Gianturco [9, 10] we are correcting the experimental integral cross sections for the angular discrimination error. We follow the procedure outlined by Kauppila et al. [19] and Kwan et al. [20] in order to take into account the effect of electro-magnetic fields in the scattering cell. Our computed integral cross sections are in good agreement with the corrected experimental data. Below collision energies of 10 eV the agreement is usually within 10 per cent. Cross sections for positron collision with gas-phase uracil have been measured by Surdutovich et al. [21] at Detroit and by Anderson et al. [22] at ANU. In Franz et al. [11] we compare our computed cross section with the experimental data from Surdutovich et al. [21]. The agreement is very poor. We did not apply any correction for the angular descrimination to the experimental data and the partial pressure of uracil in the experimental setup is not clear. The agreement is much better between our computed data and the measured data of Anderson et al. [22]. A direct comparison has not been done, because both manuscript have been published around the same time. For the DNA bases no experimental is available. For these molecules we are employing a similar model as for the other target molecules. Therefore we assume that the uncertainty of the computed data is similar to that observed for pyrimidine and THF.

The data does not include cross sections for ionization, annihilation or Positronium formation. Among the inelastic channels only rotational excitation is considered, and for uracil also vibrational excitations have been calculated.

4. Data model

Figure 1 shows the logical model of the database. The model has the following entities:

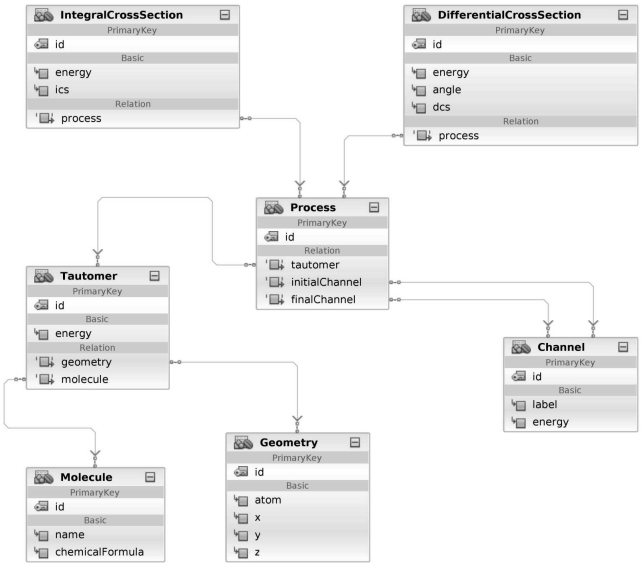

Fig. 1. Logical model of the POCOBIO database.

- **[IntegralCrossSection:]** An integral cross section is defined for a scattering process. This table contains for each process the integral cross section (ICS) for each energy. The ICS is given in units of Å^2 (this is: 10^{-20} m^2). The energy is given in units of eV.
- **[DifferentialCrossSection:]** A differential cross section is defined for a scattering process. This table contains for each process the differential cross

section (DCS) for each energy and scattering angle. The DCS is given in units of $Å^2$/sr, angles are given in degrees and energies in eV.

- **[Process:]** A process is defined for a tautomer and for an initial and final channel.
- **[Channel:]** A channel has a label and a threshold energy. The energy is given in eV and the label is a string (or VARCHAR in SQL).
- **[Molecule:]** A molecule has a name and a sum formula.
- **[Tautomer:]** A molecule can have several tautomers. Each tautomer belongs to a parent molecule and has a total energy and a geometry. The total energy is given in kJ/mol.
- **[Geometry:]** The geometry is defined by the atomic symbols and their coordinates. The coordinates are given in cartesian coordinates and are in Å.

This data model differs in several ways from data models used in the literature.

Here the entity for a tautomer contains the formation energy. These are computed automatically by most quantum chemistry software packages. We prefer formation energies over enthalpies or Gibbs free enthalpies for the tautomers, because the latter quantitaties contain also contributions from molecular vibrations and rotations. We prefer that the rotational, vibrational and electronic states are defined through the scattering channels. A thermodynamic distribution can be achieved by a weighted sum of the various cross sections.

Each scattering channel is described by a label. This label can be defined quite general and can include the rotational, vibrational and electronic state, if necessary. In the case of Positronium formation, the electronic state of Positronium can be absorbed into the label of the channel.

We do not inlude reactive scattering channels in our simple model. Therefore the tautomer (and the molecule) is the same before and after the collision. For a more general approach, like in the QDB data model of Tennyson et al. [2], the tautomer should be included in the scattering channel and should not be part of the entity process. For example the QDB data model uses an entity for a reaction, which has reactants and products. This can be compared with our entities for a process with initial and final channels.

The integral and differential cross sections are defined as tables of the relational database. Other databases like QDB are saving the cross section data in a seperate file and provide the file name and the description of the data. We have chosen this design to simplify the access and usage of the data by third parties.

5. Future developments

Currently our software for computing scattering cross sections is undergoing some major revisions and reimplementions. We have generalized our scattering codes to include complex absorbing potentials (CAPs) [23] and new correlation-polarization potentials [24]. CAPs can be used to efficiently describe channels like ionization and Positronium formation, as shown by Staszewska et al. [25], Reid and Wadehra [26, 27], and Blanco and Garcia [28, 29]. We have computed annihilation previously (see Annihilation cross sections can be calculated with the same formalism as our elastic and inelastic cross sections (see e.g. Franz and Gianturco [30]) or with CAPs, as shown by Mitroy and Ivanov [31].

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