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Positron Scattering at Thermal Energies

K. Fedus* and G.P. Karwasz
Institute of Physics, Nicolaus Copernicus University, Grudziądzka 5/7, 87-100 Toruń, Poland

Modified effective range theory in analytical form is used to extrapolate elastic cross-sections down to thermal energies for positron scattering from five atomic and molecular targets including Ar, H_2, N_2, C_6H_6 (benzene) and C_6H_{12} (c-hexane). The derived scattering parameters are then used to estimate the annihilation rates for chosen elements using a simple formula by Gribakin. We show that a combination of two simple models has a potential for accurate parameterization of coefficients describing the interaction of thermal positrons with non-polar targets of low polarizability such as Ar and N_2.

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1. Introduction

Positron annihilation in metals and semiconductors are dominated by short-range Coulomb interactions [1]; therefore, positron lifetimes in defect-free metals show values below some 200 ps [2]. This is not the case of liquids and Van der Waals solids, like hydrocarbons, where apart from long-lived components some intermediate positrons lifetimes appear at 400–500 ps range [3]. In hydrocarbons such as cyclohexane and benzene, the first lifetime component is quite short, 214 and 141 ps, respectively, see [3]. This would indicate a rather strong interaction between the molecule and the injected positrons. However, relatively few experiments for annihilation in liquids were performed; experimental values of positron lifetimes in liquefied noble gases are also quite sparse, see [4]. As a consequence, few theoretical works were done, either.

Positron scattering from materials in a gas phase allows to model the interaction between a single atom/molecule and incoming positron [5]. However, in order to deliver input data for the analysis of positron interactions in dense gases, liquids and modeling living tissues [6], cross-sections at very low (i.e. thermal) energies are also needed. These energies are usually below the typical range of electron-beam experiments (see for example [7]) and positron-swarm experiments are still to be developed (see for example [8]). In the high-energy limit positron and electron-scattering cross-sections merge [9], so electron data can serve as indication for positron modeling. This is not the case of low energies, where electron cross-sections are usually higher than positron cross-sections, even by a factor of few folds, see for example [5].

The experimental techniques developed within the last fifteen years for direct measurements of total cross-sections [10, 11] in the range of few eV range combined with a novel theoretical effective-range methodology [12–14] allows to extrapolate positron-scattering cross-section down to thermal energies. In this paper we will apply modified effective range theory (MERT) to derive elastic scattering cross-sections for thermal positron collisions with five atomic and molecular targets: Ar, H_2, N_2, C_6H_6 (benzene) and C_6H_{12} (c-hexane). On the basis of this analysis some implications for the positron annihilation rates in these materials will be discussed.

2. Experimental data

Present analysis uses a consistent set of experimental data, obtained in the past by one of the authors (G.P.K.) and colleagues with a low-energy, electrostatically-guided positron beam [10]. The essential improvement in that apparatus as compared to early setups from Wayne State University in Detroit [15] and Tokyo Yamaguchi University [16] time-of-flight experiment consists of using a new-type of positron moderator (1 μm thick W monocrystal) and narrow (1 mm diameter) slits in a relatively long (100 mm) scattering cell. The W-monocrystal moderator in the transmission geometry [17], the electrostatic optics [17, 18] and a weak guiding magnetic field working in an energy-dispersion mode [19] ensured about 180 meV energy resolution. The narrow slits ensure the angular resolution by up two orders of magnitude better than in previous techniques from Detroit and Tokyo, see Ref. [20] for discussion.

Cross-sections used here for benzene and cyclohexane have been reported in [21] and for N_2, Ar and H_2 in [22]. Unfortunately, the two earlier experiments, from Detroit [15] and Tokyo [16] used slightly modified experimental settings for different gases so the comparison between different targets is not straightforward. On the other hand, the most recent experimental technique from ANU in Canberra [11] uses very strong (500 G) magnetic field that makes complex the analysis of the scattered positrons signal. For example, in H_2O the correction for forward scattering at 1 eV is more than 200% of the directly measured value [23].

3. Modified effective range method

At large distances the interaction between the positron and an atom or a quasi-spherical molecule is dominated

*corresponding author; e-mail: kamil@fizyka.umk.pl

by the long-range polarization potential $-\alpha e^2/2r^4$, where α is the dipolar polarizability and e is the elementary charge. The Schrödinger equation for such a potential is solved exactly with the help of the Mathieu functions [24, 25]. Integral elastic (σ) cross-sections can be then calculated using the standard partial wave expansion

$$\sigma = \frac{4\pi}{k^2}\sum_{l=0}^{\infty}(2l+1)\sin^2\eta_l(k), \tag{1}$$

where k is the momentum of incident positron, l is the angular momentum quantum number and η_l is the phase shift introduced into the l-partial wave by a long-range scattering potential. The latter is obtained from the following relation [12–14]:

$$\tan\eta_l = \frac{m^2 - \tan^2\delta_l + B_l^{\%}\tan\delta_l(m^2-1)}{\tan\delta_l(1-m^2) + B_l^{\%}(1-m^2\tan^2\delta_l)}, \tag{2}$$

where $\delta_l = \pi(v - l - 1/2)/2$. Here m and v denote the energy-dependent parameters which have to be determined numerically from analytical properties of the Mathieu functions, see [12–14] for details. Another energy-dependent parameter $B_l^{\%}(k)$ related to the unknown short-range potential (non-zero at very close positron–target distance) is expanded around zero energy for each partial wave separately: $B_l^{\%}(k) \approx B_l(0) + R_l R^* k^2/2 + \ldots$, where R_l can be interpreted as the effective range for a given partial wave. Here $R^* = \sqrt{\alpha e^2\mu/\hbar^2}$ denotes a typical length scale related to the r^{-4} interaction where μ is the reduced mass of projectile/target system and $\hbar$ is the Planck constant. In the particular case of $l = 0$, $B_0(0)$ can be expressed in terms of A, the s-wave scattering length, as $B_0 = -R^*/A$. We have applied this semi-empirical model to verify that in the regime of energies for elastic scattering the leading contributions for studied targets come from s, p, and d waves, while the contribution of higher partial waves is small and can be described by taking only the leading order contribution to the phase shift

$$\tan\eta_l(k) \approx \frac{\pi\alpha k^2}{8(l-1/2)(l+1/2)(l+3/2)}, \quad \text{for } l > 2. \tag{3}$$

Consequently, substituting Eq. (2) for three first partial waves into Eq. (1) one gets relations which can be fitted to experimental cross-sections in order to determine unknown parameters of the effective range expansion.

4. Results

In Fig. 1 we present MERT extrapolations of positron low-energy total crosssection for H_2, N_2, Ar, C_6H_6, and C_6H_{12}. The corresponding parameters of the effective range expansions are given in Table I. In calculations we used experimentally determined dipole polarizabilities given in the last column of this table [26, 27]. The fittings to targets with low polarizability, that is H_2, Ar, and N_2, are relatively good and only two partial waves need to be treated accurately to reproduce experimental data in chosen energy ranges (up to 10, 4, and 2 eV for H_2, Ar, and N_2 respectively). On the other hand, the fitting procedure for benzene and c-hexane is much more tedious due to the limited number of experimental points for these materials in elastic region (i.e. below 5 eV for both molecules). Moreover, the large size of the molecules and very high polarizabilities make necessary to include the contribution of three partial waves. To reduce the degree of freedom in the fitting procedure we fixed the scattering lengths at $A = 9.52a_0$ and $13.04a_0$ for benzene and c-hexane, respectively, in order to exclude B_0 from the fitting parameters (here a_0 denotes the Bohr radius). The values of scattering lengths were chosen using $E_b = 1/(2A^2)$ (in atomic units) — the positron binding energies (80 meV and 150 meV for C_6H_{12} and C_6H_6, respectively) that are determined experimentally for both molecules, see [28] and references therein for more details. Here we use the fact that in large molecules, such as benzene and c-hexane, the formation of weak positron bound state is present.

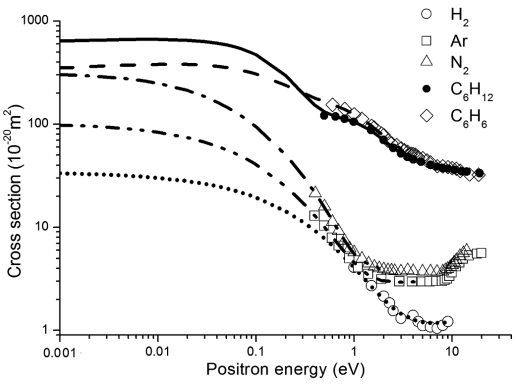

Fig. 1. Present MERT extrapolation towards zero-energy of positron low-energy total cross-sections H_2, Ar, N_2, C_6H_6, and C_6H_{12}. Experimental data are from Karwasz et al. [21, 22].

TABLE I

Parameters of the effective range expansion for positron scattering from studied targets: $A = -R^*/B_0$, B_1, B_2, R_1 and R_2. In the last column the dipole polarizability values used in calculations are given [26, 27].

	A [a]	R_0 [a_0]	B_1	R_1 [a_0]	B_2	R_2 [a_0]	α [a_0^3]
H_2	−3.13	−0.12	−1.99	−0.90	–	–	5.314
Ar	−5.52	0.94	−4.34	4.76	–	–	11.23
N_2	−9.23	2.56	−22.67	69.04	–	–	11.54
C_6H_{12}	13.04*	5.51	1.16	−0.05	−0.18	0.36	72.50
C_6H_6	9.52*	1.77	−0.23	1.05	−0.29	0.47	73.44

* Fixed on the basis of experimentally determined positron binding energies [28].

To obtain some information about positron annihilation rate (λ) using derived elastic scattering cross-section (σ) we used the formula introduced by Gribakin [29] for positron direct annihilation at very low (thermal) energies

$$Z_{\text{eff}} = 4\pi\rho_e\delta R\left(R^2 + \frac{\sigma}{4\pi} + 2R\text{Re}\{f_0\}\right), \tag{4}$$

where $Z_{\text{eff}} = \lambda/(\pi r_0^2 cn)$ with r_0 denoting the classical

electron radius, c — the speed of light and n — the number density of target (atom or molecule). Here ρ_e is the electron density in the annihilation range, δR is thickness of a thin shell enclosing the target system (for more details see Ref. [29]), R is the radius of the target and f_0 is the spherically symmetric part of the scattered amplitude. The latter can be expressed in a terms of the s-wave phase shift η_0 as $\mathrm{Re}\{f_0\} = \sin(2\eta_0)/(2k)$. Equation (4) is very useful for a rough estimation of the annihilation rate, though the electron density ρ_e and the coefficient δR can be found only by comparison with some numerical data.

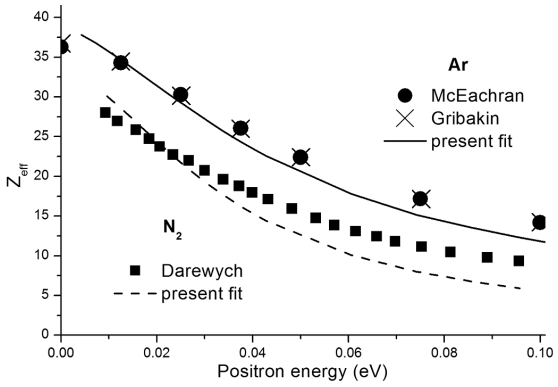

Fig. 2. Annihilation Z_{eff} for argon (Ar) and molecular nitrogen (N_2) as a function of positron energy. The data are from McEachran et al. [30] (circles), Gribakin [29] (open squares) and Darewych and Baille [31]. The solid and dashed lines present the prediction of Eq. (4) using the scattering cross-sections and phase-shifts calculated in this papers, and considered $R = R^*$ and the prefactor $4\pi\rho_e\delta R$ as then only fitting parameter.

In Fig. 2 we present the fit of Eq. (4) using the elastic scattering cross-sections and the s-wave phase-shifts derived in this work to *ab initio* calculations of Z_{eff} for argon by McEachran et al. [30] and nitrogen by Darewych and Baille [31]. In particular, the dataset of McEachran is a reliable reference because it was proved to be in very good agreement with experiment by Kurz et al. [32]. As the radius of the atom we used a typical length scale related to r^{-4} interaction, i.e. $R = R^* = 3.35a_0$ and $3.40a_0$ for argon and molecular nitrogen, respectively. The only parameter determined by the fitting procedure was the prefactor in Eq. (4), i.e. $4\pi\rho_e\delta R = 0.51$ and 0.21 for argon and molecular nitrogen, respectively. For comparison purposes the accurate *ab initio* calculations of Gribakin [29] are also shown in Fig. 2.

We can see that Eq. (4) combined with our MERT extrapolation reproduce very well the argon and nitrogen annihilation coefficients obtained at thermal energies by other theories [29–31]. The usefulness of Eq. (4) has been already verified for noble gases and C_2H_4 molecule in Ref. [29] but with the use of two fitting parameters, $4\pi\rho_e\delta R$ and R.

In Table II we compare the present estimates of two latter factors with results obtained by Gribakin [29] for

TABLE II

Scattering lengths and fitting parameters for the relation between Z_{eff} and σ [Eq. (4)].

Atom or molecule	A [a_0]	R [a_0]	$4\pi\rho_e\delta R$ [a.u.]
Ar*	−5.52	3.35	0.51
N_2^*	−9.23	3.40	0.21
He†	−0.52	3.90	0.21
Ne†	−0.61	5.00	0.23
Ar†	−5.30	4.30	0.42
Kr†	−10.4	4.20	0.41
Xe†	−45.3	4.20	0.41

* present estimation; † Gribakin [29]

other atomic systems. In particular, note that all presented values of $4\pi\rho_e\delta R$ are pretty consistent with simple estimates of this parameter using positronium density and high-quality atomic calculations as reported in Ref. [28] and giving $4\pi\rho_e\delta R \approx 0.5$ and 0.66, respectively. It is expected that the similar results should be obtained for other small molecules with low polarizability, also including H_2 (see for example [33]).

The formula of Gribakin (Eq. (4)) does not account for the effect of resonant annihilation due to the formation of bound states with the positron [29]. Hence, the same conclusions about $Z_{\mathrm{eff}}(k)$ tendency as derived above for Ar and N_2 cannot be assigned to C_6H_6 and C_6H_{12} with the same level of confidence. Moreover, non negligible quadrupole moments can introduce some important modifications of r^{-4} interaction. Consequently, it is difficult to get reliable picture of positron–large molecule interaction at thermal energies using present semi-empirical approach and available experimental data. For example, higher elastic cross-sections at thermal energies obtained for c-hexane in our MERT calculations indicate stronger interaction of positron with this molecule in a gas phase and presumably also in the liquid phase. As a result, the shorter positron lifetime (higher annihilation rate) is expected in C_6H_{12}. However, this is in contradiction to experimental results in liquids [3]. On the other hand, we found that when applying unconstrained fits for all parameters including scattering lengths, MERT always predicts higher cross-sections for C_6H_6 in the zero energy limit. However in this case the determined scattering lengths are not in agreement with experimentally measured positron binding energies [28]. Nevertheless, despite such ambiguity, the final verification on the applicability of the presented model to large molecules will come when experimental data of collisional coefficients at thermal energies will be available or some general consensus on *ab initio* theoretical results will be achieved.

5. Conclusions

We showed that a simple semi-empirical modified effective range analysis (MERT) can be used to predict positron cross-sections down to thermal energies. Moreover, we showed that the combination of MERT with a

simple formula by Gribakin [29] is a potential tool for a parameterization of positron annihilation coefficients characterizing the interaction with no-polar targets of low polarizability, such as noble gases and some simple molecules (N_2). In any case, further measurements down to zero energy are indispensible in order to apply both models with more accuracy for parameterization of interaction coefficients.

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