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Positron scattering in helium: Virtual-positronium resonances

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Abstract

Total cross sections for positron scattering in helium and argon were measured at 0.5–25 eV with a new spectrometer from Trento University, with a good angular resolution (3.1×10^{-4} sr). Data in argon fall in-between earlier measurements; those for He agree well with the previous above the positronium formation thresholds but are up to 50% higher at low energies. Data for He show four resonant-like structures, one at the positronium threshold formation (17.8 eV) predicted by Van Reeth and Humberston [J. Phys. B 32 (1999) L103], another at about 6.8 eV and two very prominent centred at 1.6 eV and 2.2 eV. Following Gribakin and King [J. Phys. B 27 (1994) 2639] the two latter could be signs of virtual-positronium formation in helium.

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Keywords: Positron scattering

1. Introduction

The existence of chemical compounds between matter and anti-matter, for example a hydrogen/anti-hydrogen molecule, is subject to intense studies [1]. A bound state between positronium (Ps) and atomic hydrogen has been observed in positron scattering on methane [2]. Possibilities of transient states (resonances) between atoms and positrons have been suggested for several targets but not verified experimentally yet [3,4]. Recently, narrow peaks in positron annihilation have been observed below thresholds for vibrational excitation in some heavier hydrocarbons [5], indicating resonant processes.

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Helium was a milestone of electron-scattering spectroscopy, being the first atomic target in which a resonant state was observed [6]; this structure shows up in the forward-angle differential elastic cross sections slightly below (at 19.37 eV) the threshold for the lowest-level electronic excitation as a sharp minimum of about 0.2 eV width, followed by a sharp maximum. Subsequently, these so-called Feshbach resonances have been discovered in numerous atoms and molecules (see review [7]).

Total cross sections (TCS) for positron scattering on helium at energies below 30 eV were subject to numerous experimental investigations [8–14], see review by Kauppila and Stein [15]. The most systematic studies of positron scattering were performed by the Detroit group. For He measurements at 0.3–31 eV they used a proton-irradiated boron target emitting slow positrons with the declared energy distribution of 0.1 eV and a long (109 cm), curved scattering cell with a longitudinal guiding magnetic field [11]. They obtained extremely low values of TCS in He (0.05–$0.06 \times 10^{-20}\,\mathrm{m}^2$ at 1–3 eV), much lower than the minimum in molecular and in atomic hydrogen (about $0.8 \times 10^{-20}\,\mathrm{m}^2$ at 3–5 eV in both targets [16]). The more recent data of Mizogawa et al. [14] are at low energies (1–5 eV) higher than those of Stein et al. [11] by 15–30%.

If compared to other noble gases, Ar (see Fig. 1), Kr, Xe or molecular targets, like N_2, CO_2, SF_6, CH_4, see [15], helium shows an interesting feature. In all the gases cited, the TCS rises towards the zero energy but at energies above 1–2 eV remains practically constant, up to the threshold for the Ps formation, see present data for argon in Fig. 1. In helium, according to measurements of Stein et al. [11] the TCS falls in the 0.3–1.2 eV range and then, in the range 1.2–2.7 eV, remains almost constant. Above 3 eV, much below the Ps formation threshold (17.8 eV) the TCS starts to rise, reaching a kind of maximum just at the threshold for Ps formation, see Fig. 2. Very few targets (only Ne and O_2) behave in this way, see [15].

In several theoretical works, the possibility of formation of the virtual Ps in He below the threshold for the free Ps formation (17.8 eV) has been stressed. Gribakin and King [17] using many-body perturbation theory, pointed out that the inclusion of the virtual Ps state is needed to reproduce elastic cross sections at low energies, see Fig. 3. The virtual Ps would be a temporary positronium atom formed between the incoming positron and an atomic electron. Some analogy can be found with resonances in electron scattering, where an incoming electron forms a temporary negative ion with the atom.

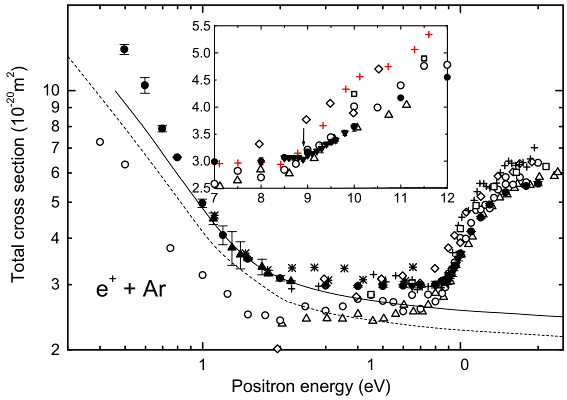

Fig. 1. Experimental and theoretical total cross sections for positron scattering in argon. Experiments: △, [32]; ✳, [13]; ○, [33]; ◇, [9]; +, [30]; □, [31]; present, full symbols (●, ▲ and ▼ correspond to different measurement sessions); the arrow indicates free Ps formation threshold. Theory: ----, [35]; —, [34].

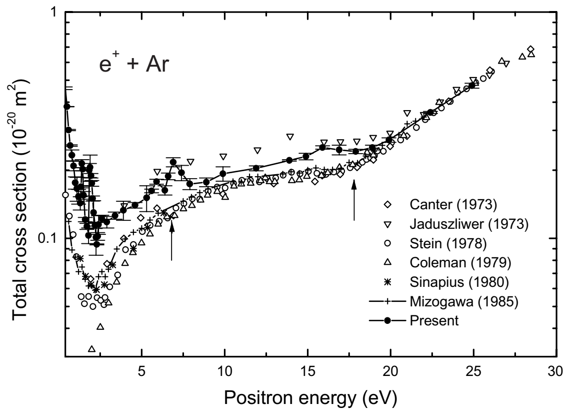

Fig. 2. Comparison of present data for helium with previous experiments. Symbols like in Fig. 1, apart from inverted triangles, [10]; ○, [11]; △, [12]; —+—, [14]. Early data of Costello et al. [8], higher than all other sets, are not shown for clarity.

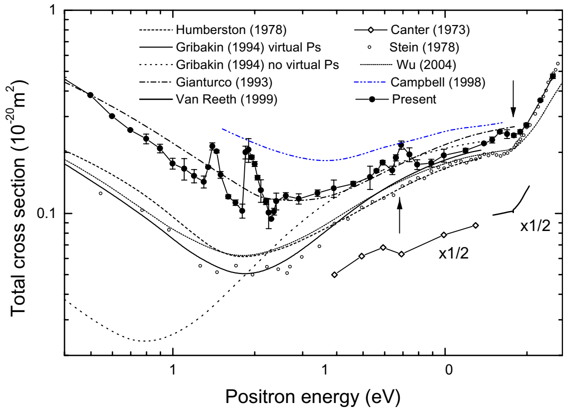

Fig. 3. Comparison of present data for helium with theory and some previous experiments. Symbols for experiments are the same like in Fig. 1. Theory: ----, [18]; —, [17] with virtual Ps channel; dotted line, [17] without virtual Ps channel; dot–dash line, [34] density functional; dash–dot–dot line, [38], R-matrix without Ps channels; light dotted line, convergent close coupling, [37]; heavy line at 17–20 eV, Ps formation resonance from [19].

Gribakin and King showed that if no virtual Ps formation is allowed, the TCS does show a similar shape to the average of the experimental data but wrong absolute values – a too deep minimum and at a lower energy. Results of [17] agree with variational calculations of Humberston [18].

A second theoretical indication for possible resonances in $e^+ + He$ scattering are the works of Van Reeth and Humberston [19] who predicted resonant structures in both elastic and Ps formation channels for atomic hydrogen and helium, see Fig. 3. They stressed the importance of virtual Ps formation not only at the Ps threshold but over a wider energy range – "the virtual Ps formation process generates a more attractive effective positron–atom potential, the positronium sticking more to the ionized target and making the phase shift more positive". Resonant structures in experimental elastic cross sections at the Ps threshold were evidenced for Ar, Kr and Xe, but no in Ne (no data for He are available) [20]. Finally, the existence of a metastable bound state between positron and the excited helium He 1s2s ^{3}S atom with the binding energy in para-state of −0.016 eV and the lifetime of 176 ps (compared to 125 ps of free para-Ps) has been predicted recently by Ryzhikh and Mitroy [21]. This state can be considered as a Ps atom orbiting a He^+1s core at large distances.

We measured TCS in helium and argon with a new apparatus from Trento University, characterized by small apertures of the scattering cell (1 mm in diameter) and low guiding magnetic field (10 Ga), both essential for good angular resolution, and using a new type, tungsten monocrystal moderator with possibly good energy resolution. The good beam stability and counting rate allowed to evidence an almost constant dependence of the cross section in argon on energy at 3–9 eV but in helium some sharp, resonant-like structures in TCS at 1.4–2.6 eV.

2. Experimental

Present measurements have been performed with a new, electrostatically-guided positron beam at Trento University. The apparatus has been described in detail in our previous paper [22], where also detailed drawings were given. However, compared to the description in [22] some essential simplifications were done, as discussed below. The construction is based on our previous experience with a variable-energy positron beam for studies of defects in solid state via positron annihilation techniques [23]. Some test measurements in molecular nitrogen and benzene are discussed in [24].

The positron source is the ^{22}Na isotope. The apparatus uses exclusively electrostatic lenses in the first part of optics, a 90° spherical electrostatic analyser, and both electrostatic focusing and the longitudinal magnetic field in the second part of optics and in the scattering cell region, see Fig. 1. A significant improvement compared to previous experiments in atomic physics [8–14] is our positron moderator and background vacuum conditions (4×10^{-9} mbar). We use tungsten 1 μm thick monocrystal from Aarhus University [25], annealed in situ in UHV (10^{-8} mbar) conditions by a telefocus electron gun. The final temperature of annealing is difficult to estimate but the film gets "brilliant-white" for a dozen seconds, exceeding the temperature of commercial tungsten bulb lamps (2500 K).

The energy selection of the beam is performed in several stages. First, compared to magnetically guided beam [26] the present apparatus selects only a fraction ($\pm 15°$) of positrons emitted from the moderator. Secondly, some energy and/or geometry selection is performed by the 90° bend; the FWHM of this selector is 1.6 eV.

The scattering cell made of copper–nickel alloy is 10 cm long and has 1 mm diameter entrance and exit apertures (3.1×10^{-4} sr is the geometrical angular resolution). Another 5 mm diameter aperture is placed 3 cm in front of the cell. A longitudinal magnetic field (about 10 G) is slightly ($\pm 10\%$) adjustable, in order to obtain an integer number of gyrations of positrons inside the scattering cell (i.e. 3 gyrations for 2.4 eV positrons). Measuring the vibrational structure in the total electron scattering on molecular nitrogen we have evaluated the energy resolution (FWHM) of the scattering cell assembly as better than 130 meV [27].

A third factor determining the resolution of the apparatus is the width of energy spectra of positrons emitted from the moderator. Fischer et al. [28], using a hemispherical electrostatic analyzer with a 20 meV resolution, reported the energy width as low as 42–50 meV for positrons re-emitted backwards from the W (1 1 0) monocrystal. Amarenda et al. [29] reported a wider $\approx$0.25 eV FWHM energy distribution from the W (1 0 0) foil annealed in situ but the energy resolution of their determination is unclear – they used simply a retarding potential analyzer.

We are not aware of any sharp structures in positron scattering cross sections which would allow to determine directly the energy resolution of our beam. Taking into considerations all components, the energy resolution of the present apparatus is probably 130 meV but it could be as good as 50 meV, if determined by the energy distribution of positrons emitted from the W-monocrystal and the optics (the bend, the magnetic lens in the scattering cell) cutting-off any tails of this distribution.

Compared to the previous descriptions [22] in the present mode of operation the electrostatic potentials in the second part of optics are inverted, from accelerating ones (needed for a remoderation stage) to decelerating ones (needed for the scattering experiment without a remoderator). With this modification the beam can be still well focused at the entrance of the scattering cell, but due to Helmholtz–Lagrange's principle a significant part of the beam intensity is lost – only 1/100 positrons are injected into the scattering cell with "useful" angles, see Fig. 2. The beam intensity remains 20–100 e^+/s in the whole 0.4–25 eV energy range. Note that this is much higher than in measurements of Mizogawa et al. [14], who obtained 0.4 e^+/s with the 13 Ga magnetic field. We also do not use at present the concept of time-of-flight [22], what simplifies the construction but creates some difficulty in the determination of the energy scale bias.

The shift of the energy scale (the sum of the moderator positive work function and contact potentials) has been estimated from the Ps formation threshold in N_2 [24] and Ar, see Fig. 1, and amounts to $+2.4 \pm 0.1$ eV. This means that below the 2.4 eV collision energy the apparatus works with decelerating voltages and its energy resolution can be better than at higher energies.

The cross sections are determined by the de Beer–Lambert attenuation formula,

$$I = I_0 \exp(-pl\sigma/kT), \tag{1}$$

where l is the length of the scattering cell, p is the gas pressure, T is temperature of the gas and k is Boltzmann's constant. Pressure was determined by an absolute capacitance membrane Baratron MKS 628B meter with better than 1% precision. The main source of potential systematic error is the pressure determination, including a possible thermal transpiration (+3.5%). Therefore, the measurements were performed in long series of constant pressure, changing the projectile energy. The experimental points shown in Figs. 1–4 are mean values of 6–10 runs, each performed as 20 values of 10 s count accumulation with gas on and off. Error bars shown correspond to one standard deviation of the mean value. Additional runs were performed with different settings between 1.2 and 2.5 eV.

Helium gas was AirLiquid, quality Alphagas1 99.999% with H_2O (3 ppm), O_2 (2 ppm) and CH_4 (0.5 ppm) main contaminants; the purity was checked independently with a mass spectrometer. The gas line was heated under vacuum before connecting He and the first doses of He were used for

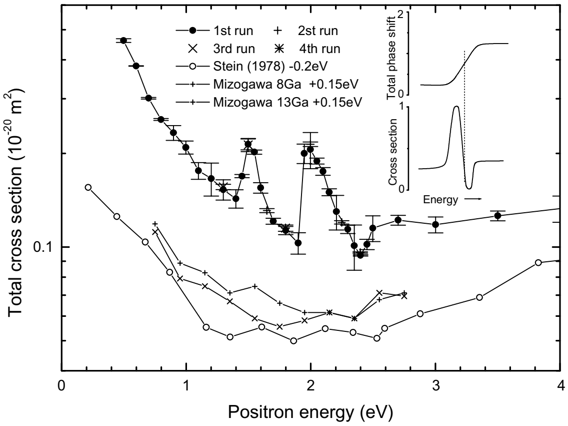

Fig. 4. Particular of the low energy range in He, present data. Different runs are shown separately. For sake of comparison the data of Stein et al. [11] have been shifted by −0.2 eV and those of Mizogawa et al. [14] performed with 8 Ga magnetic field by +0.15 eV. The inset shows a resonant variation of the phase shift and the corresponding structure in the cross section, see [7].

flushing the line. During measurements the gas line was kept overpressure compared to the atmosphere.

3. Experimental data

Our data for argon are shown in Fig. 1 – they agree well with other determinations: in the energy range between 2 and 10 eV they almost coincide with the data of Charlton et al. [30], Canter et al. [9] and more recent data from the Detroit laboratory [31] and are slightly lower than those of Sinapius et al. [13] but are by about 20% higher than those of Coleman et al. [32] and earlier data from the Detroit laboratory [33]. Above the Ps thresholds our data agree best with those of Kauppila et al. [33] and Coleman et al. [32]. Below 2 eV present data agree better with recent theoretical elastic cross sections [34,35] than the data of Kauppila et al. [33]. In spite of numerical differences, all measurements indicate a constant cross section in argon between about 2 eV and the free Ps threshold.

In helium, present measurements above the Ps threshold agree very well with all measurements presented in Fig. 2 (only the early data of [8] have been excluded). In the range 4–18 eV present data are lower by 10–20% than measurements of Jaduszliwer and Paul [10] but higher by a similar value than the rest of experiments [9,11–14]. Below 3 eV present data are higher than those of Stein et al. [11] by almost 50%. The reason is not clear for us – the same sign difference exists for argon, see Fig. 1 and molecular nitrogen, see [24]. The most probably it is related to the angular resolution: in TCS measurements some of the scattered projectiles, these within the aperture of the scattering cell exit, are counted as non-scattered and lower the measured TCS value. The use of the magnetic field worsens the error – all projectiles scattered with the transverse energy lower than a certain value are recaptured by the magnetic field and guided to the collector. With typical values used in positron experiments, 13 Ga of the magnetic field [14] and 2.6 mm radius of scattering cell exit apertures [11] this transverse energy equals to 1 eV. This means that at 2 eV collision energy all positrons scattered below the angle of 30° are counted as non-scattered; at 1 eV collision energy this angle amounts to as much as 90°. In order to calculate the correction in TCS, detailed differential cross sections must be known. In nitrogen, using recent detailed theoretical data [36], we have calculated [37] the possible correction due to this effect, assuming 4 mm radius apertures and 9 Ga field. This correction amounts to as much as a factor of two at 1.4 eV, a factor of three at 1 eV, and a factor of seven at 0.5 eV where the scattering is particularly forward centred. For our apparatus the correction in nitrogen is as low as 4% at 0.5 eV. In argon and helium we are not aware of such detailed differential cross sections down to zero energy; the value of the magnetic field used is not given in [11] either. The fact that our data differ from those from the Detroit lab less in argon (30% at 1 eV) than in He (by a factor of two at the same energy) would indicate a more forward-peaked scattering in He than in Ar.

Present data in He agree very well with calculations of De Fazio et al. [38] in the density-functional model with a multiterm polarizability and spin orientations included explicitly, see Fig. 3. Present data are higher than recent convergent close coupling data [39] and lower than the R-matrix results [40].

4. Virtual-positronium resonances in He

The most interesting feature of present measurements is the presence of four resonance-like structures, see Figs. 3 and 4. The highest-energy structure is placed just below the positron threshold and would correspond to that predicted by Van Reeth and Humberston [19]. The amplitude of the measured structure agrees roughly with the theoretical value of 7% but both present data and those of Mizogawa et al. [14] show an inverted shape, see Fig. 3. Note that some lowering of the cross section just before the Ps formation threshold was seen also in data of Stein et al. [11].

The most prominent are two structures, with thresholds at 1.4 eV and 2.0 eV, see Fig. 4. At present, we lack both theoretical and other experimental confirmation for them, relying only on some indications. Analysing the only other experimental data extending below 2 eV, we note that the data of Stein et al. [11] between 1.2 and 3 eV show somewhat bigger fluctuations than at higher energies; if shifting their data by −0.2 eV (compatible with the combined energy scale uncertainty in our and their experiment), maxima of these "fluctuations" would coincide with ours but are much lower, see Fig. 4.

Mizogawa et al.'s data are given for energy ranges by 0.2 eV, i.e. 0.5–0.7 eV, 0.7–0.9 eV. In Fig. 4 we attributed their values to the mid of the ranges; a shift by +0.15 eV is again experimentally plausible and would correspond to the mismatch in their measured position of the electron–He resonance (19.2 eV) with the literature value (19.36 eV). Note from Fig. 4 that Stein et al's [11] shifted data would agree in shape with Mizogawa et al.'s [14] TCS obtained with 8 Ga field, but are by 15–30% lower. Then, Mizogawa et al.'s data obtained with 13 Ga magnetic field coincide with their 8 Ga data, being only slightly lower, apart from 1.5–1.7 eV (the region of first resonant peak) where they departure down. To make some hypothesis on the lacking observation of resonant structures in earlier measurements one should discuss two experimental features – the energy resolution and the forward-scattering error.

The apparatus of Mizogawa et al.'s [14] was characterized by a better angular resolution than that of Stein et al. [11] but by a worse energy resolutions. The energy spectrum of positrons emitted from bulky tungsten moderators (i.e. in the form of mesh or Venetian blinds [14]) is a continuous distribution up to 2.4 eV, see [28,29]. The use of the magnetic field performs some energy selection, as was observed also by us [27] but for sure the energy resolution of the Mizogawa et al.'s set-up is worse than 0.1 eV declared by Stein et al. [11]. On the other hand, the curved solenoid [11] requires higher magnetic field values than a linear set-up [14], worsening the angular distribution. Once again, differential cross sections at the resonance should be known to evaluate the possible error. We note only that in helium electron scattering, the 19.36 eV resonance has a well pronounced, up-and-down shape for forward-scattering angles, up to 72°, see [7]. At 90° the first peak disappears. In argon, the $3p^5(^2P_{3/2,\ 1/2})4s^2(^1S)$ resonance at 11.1/11.3 eV appears in forward differential cross sections as two sharp maxima, up to the angle of 120° and then, at 140° has a totally inverted shape [41]. If resonant positron–helium scattering is forward centred, the structure can be overlooked in measurements with a poor angular resolution: contributions from backward angles can cancel the structure seen in forward scattering.

The two low energy resonances, to our knowledge, are not predicted by the theory. Following the idea of Gribakin and King [17] about formation of virtual positronium, the question arises if this formation is subject to a threshold, like the free Ps formation. If so, the two resonant structures could indicate thresholds for virtual Ps formation in two possible spin states. Unfortunately, theories do not agree on energies and very existence of the virtual positronium in He and bound states positron- helium, see the paper by Mitroy and Novikov [42].

Note from the insert in Fig. 4 that the two structures, with a maximum followed by a minimum, would indicate a resonant, $+\pi$ change of the phase shift superimposed on a small, positive phase shift from the potential (i.e. non resonant) scattering. Generally, as discussed by Schulz [7], the resonances could appear in cross sections as single maxima or minima or combinations of them. Different theories, see discussion in [38], predict at 3 eV ($k = 0.38$ a.u.) the s-wave phase shift of about +0.05 rad descending with k while the p-wave phaseshift is about +0.03 rad rising with k. Therefore, the phase shift due to potential scattering is comprised between 0 and $\pi/2$ and the shape of the structures with the $+\pi$ resonant shift should be a maximum followed by a minimum [7], see the inset in Fig. 4.

The fourth structure observe in our cross section is centred around the free Ps binding energy, 6.8 eV; we have no any tentative explanation for its nature. An abrupt drop of the cross section between 5 eV and 6 eV was seen in the data of Canter [9], see Fig. 3; measurements of Mizogawa et al. [14] unfortunately lack between 6.25 and 7.25 eV.

Neither of calculations of the $e^+ + \mathrm{He}$ scattering process predict resonances at energies much below free-positronium formation threshold. Mitroy and Novikov [42] predicted bound states for the structures (m^{Z+}, e^-, e^-, e^+) but argued that for $Z = 2$ it would be stable only with the mass $m^{2+}/m_e \leqslant 0.68$.

A rough diagonalization [43] of the ab initio hamiltonian for the total spin quantum number equal to 1/2 , with the basis of 768 explicitly correlated Gaussian spatial functions (optimized for the lowest quartet state [1]) gives the following eigenvalues below Ps formation threshold, corresponding to positron energies of 0.544 eV, 2.451 eV, 6.428 eV, 12.722 and 17.773 eV. The last state is degenerated with the optimized quartet state and its existence has been predicted earlier by Ryzhikh and Mitroy [3]. These theoretical values could correspond approximately to the structures seen in TCS: that at 0.54 eV would be superimposed on a strong potential-scattering background, the structure at 12.7 eV is not to be excluded but more measurements are needed. This would leave only the 1.6 eV structure with no corresponding e^+–He eigenvalue.

5. Conclusions

In conclusion, TCS at 0.5–25 eV have been obtained with a new positron beam from Trento University for argon and helium. The machine works in UHV conditions, uses in situ W-monocrystal moderator in the transmission geometry, is characterized by a good angular resolution and a high counting rate. Our data in Ar lay in-between other experiments. Present data in He agree reasonably well in the 10–25 eV range but below 3 eV show two very prominent and partially overlapping resonant structures. As far as resonant structures centred slightly below the free Ps formation threshold (17.8 eV) and around 6.8 eV seem to have some previous experimental evidence; the structures below 3 eV were not seen before.

To exclude possible experimental artefacts we repeated measurements in different conditions of pressure and magnetic field: the data are reproducible within the stability margin of the energy supplier. To exclude impurities, we have analysed the gas with mass spectrometer: it does not contain molecular impurities (like N_2 or H_2) which would produce some structures, for example due to vibrational excitation. On the other hand, no partial cross section show so complex energy dependence. We have also measured TCS in N_2 with the 0.1–0.2 eV step in the 1–3 eV range and the cross sections is smooth, descending with energy [37].

We stress that the shape of the structure is exactly opposite to this in electron scattering in He seen at forward angles. For electrons, the inner atomic potential is attractive, so the wave phase shift is negative; for positron scattering the potential is repulsive, what would produce a positive shift. If compared with the theoretical phase shifts for potential scattering, the shape of the structures would be exactly as presently observed.

However, the structures are so sharp, that they indicate the energy resolution of the apparatus better than 150 meV. We argued from works on W-moderators in transmission geometry that it could be as good as 50 meV. For sure, present apparatus has the best angular resolution ever used; the geometrical resolution is better at present by a factor of 100 as compared to some other very recent positron experiments [44].

Resonant structures in electron–atoms scattering are seen only at thresholds for inelastic processes, like the electronic excitation. At 1.4–2.6 eV no inelastic thresholds occur in He. However, if temporary attachment of positron to helium atom is possible, as postulated by theory [17], this process can be subject to a threshold. The width of the two low-lying structures would indicate the lifetime of the $He^+ + Ps$ structure to be 10^{-15} s, so quite long in atomic scale. Static calculations would yield correct eigen-energy values of $He^+ + Ps$ but fail in reproducing cross sections [43].

Existence of resonant structures needs to be confirmed both in measurements of TCS in new-type, high-resolution set-ups, like that of Surko and collaborators [4] as well as in measurements of differential cross sections below 5 eV. From the theoretical point of view, more works on virtual positronium would be needed. Note that both works assuming existence of virtual positronium explicitly [17], as well as works not using this concept [34] predict correctly measured TCS values, apart from the resonant structure. Also works predicting resonant structures [18] do not differ much from the experimental data.

The simple, (α, e^-, e^-, e^+) four body system remains puzzling.

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