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LETTER TO THE EDITOR

Search for resonances in the scattering of low-energy positrons from atoms and molecules

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Abstract

In a search for quasi-bound states, or resonances, high-resolution measurements (ΔE ~25 meV) of the total positron scattering cross sections have been carried out in the energy region of the first electronically excited states of H_2, N_2, CO and Ar. In the case of H_2, a recent calculation by Varella *et al* predicts such a resonance in the total elastic cross section near the excitation threshold for the B $^1\Sigma_u^+$ state. We find no experimental evidence for the existence of this resonance and also find similar negative results for the other atomic and molecular targets.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Low-energy electron scattering cross section measurements and theoretical calculations are replete with examples of the formation of quasi-bound negative ions, or resonances (Buckman and Clark 1994). Indeed most atoms and molecules exhibit negative-ion resonance features at energies below the first ionization potential. The primary mechanisms for the formation of these complexes arise either when the projectile electron is temporarily bound due to the combination of a repulsive angular momentum barrier and the attractive polarization potential to enable the formation of a 'shape resonance' or when a doubly excited complex of the negative ion occurs just below the energy of a neutral excited state, in which case a Feshbach resonance occurs. The former is usually, but by no means exclusively, associated with the ground state of the system and decays very strongly into it by autodetachment. The latter, on the other hand, lies below its 'parent' state in energy and must decay by autodetachment into other lower-lying excited states, or into the entrance channel. As a result, shape resonances have short lifetimes

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($\sim 10^{-15}$–10^{-14} s) and are generally very broad ($\sim$ several hundred meV) whilst Feshbach resonances typically exhibit longer lifetimes and narrower energy widths ($\sim$1–10 meV).

There are now many accurate calculations for bound states of positrons with atoms (see e.g. Ryzhikh and Mitroy 1997, Mitroy *et al* 1999) and the existence of positron–molecule bound states (or quasi-bound resonances) has been postulated to explain the anomalously high annihilation rates observed in a variety of molecules (Smith and Paul 1970, Surko *et al* 1988, Iwata *et al* 2000, Gribakin 2000). However, there have been no direct experimental observations of resonances in positron scattering cross sections for either atoms or molecules. Such features have long been sought in positron scattering cross sections but, until now, a combination of nature and technology has more or less mitigated against their observation. For example, the repulsive nature of the static Coulomb potential for positron scattering results in there being little likelihood that the weak potential barrier needed to form a shape resonance will exist for positron interactions with either atoms or molecules. It is also the case that the majority of positron scattering experiments to date have been performed with reasonably poor energy resolution (typically $\Delta E \geqslant 0.5$ eV) and, as a result, any sharp and/or weak features are unlikely to have been observed. One notable exception to this is a study by Stein *et al* (1981), where they searched, unsuccessfully, for resonances in the total cross section for positron scattering from Ar, He and H_2 with an energy resolution of $\sim$100 meV.

On the other hand, there have been a number of theoretical predictions of resonances in positron scattering on light atomic systems such as H and He. In particular there have been many predictions of the existence of Feshbach resonances near the $n = 2$ excited states of atomic hydrogen. These were first proposed by Mittleman (1966) and have since been investigated using a wide variety of techniques such as close-coupling, complex-coordinate rotation, R-matrix and hyperspherical close-coupling approaches. A summary of this paper can be found in the article by Gien (1996). The essential features of the most sophisticated of these calculations are the identification of a number of resonances in positron scattering from atomic hydrogen, which lie below the thresholds for excitation of the H atom to $n = 2, 3, 4$. Resonances are also predicted in the positronium formation channel near the Ps($n = 2$) threshold. While there is some variation in the predicted widths of these features it appears that most are less than 1 meV wide. Thus it is uncertain as to whether the resonance cross sections are large enough to render them visible, even in a high-resolution ($\sim$25 meV) positron scattering experiment. Coupled-state calculations (Campbell *et al* 1998) have revealed similar structures near the He ($2\,^{3,1}$S) thresholds and the Ps($n = 2$) threshold for positron scattering from He. There is no reason to believe that such features would be limited to helium, and so similar predictions might be expected for other atomic or molecular species.

One further motivation for the present study comes from a very recent calculation by Varella *et al* (2001), who have predicted the existence of a sharp Feshbach resonance in elastic positron scattering from H_2. They used the Schwinger variational technique in a fixed-nucleus calculation which includes the static and polarization interactions. The polarization effects were accounted for by allowing for single excitations of the $(N+1)$-particle system. Varella *et al* found a Feshbach resonance at an energy just below the B $^1\Sigma_u^+$ state threshold. The parentage of this resonance is a mixture of two of the lowest-lying singlet excited states, the B $^1\Sigma_u^+$ and E $^1\Sigma_g^+$ states. In this calculation the energies of these states, calculated from Hartree–Fock orbitals, are 12.75 and 13.14 eV, whereas the actual spectroscopic values of the corresponding ground vibrational levels in each are 11.19 and 12.296 eV. The predicted energy for the resonance is 12.63 eV, which is 120 meV below the calculated threshold for the B $^1\Sigma_u^+$ excitation. The predicted width is 8 meV and the predicted contribution to the integral elastic scattering cross section is about a factor of 20 larger than the non-resonant cross section, resulting in an 8 meV wide structure with a peak resonant cross section of $\sim 13\ a_0^2$ (Varella *et al* 2001).

We have undertaken a search, using high-energy resolution ($\Delta E \sim$25 meV FWHM), for resonances in the total positron scattering cross sections for the diatomic molecules H_2, N_2 and CO and for the Ar atom, in the energy region of the lowest-lying excited state thresholds. One rationale for such a search is that the energy dependence of the total cross section can be measured with the greatest sensitivity, and there are electronic excited states that may act as parents for resonance formation in this energy region.

2. Experimental apparatus and techniques

The apparatus and experimental techniques used for these studies have been described in detail elsewhere (Gilbert *et al* 1997, 1999, 2000, Sullivan *et al* 2001a). Briefly, a buffer-gas positron accumulator is used to trap and cool positrons emitted from a solid-neon-moderated ^{22}Na source. Collisions with molecular nitrogen and carbon tetrafluoride molecules provide energy loss mechanisms (electronic and rovibrational excitation) for the positron trapping and cooling. The positrons are confined using an electrostatic potential well in the presence of a magnetic field of $\sim$0.1 T. By carefully manipulating the depth of the well, a pulsed beam of positrons can be released from the trap with both a well defined energy and a small energy spread (e.g. $\leqslant$25 meV). In practice, for the experiments described here, space charge considerations limit the pulses to about 3×10^4 positrons each. The pulse width is $\sim$1 μs and the repetition rate is typically 4 Hz. The pulsed positron beam is then passed through a collision cell containing the gas under study. The gas pressure in the collision cell is typically in the range 0.1–0.5 mTorr while the pressure elsewhere in the system is maintained a factor of 500 lower by differential pumping. After passing through the collision cell the positrons are guided through a retarding potential analyser (RPA). Detection is achieved by measuring the 511 keV annihilation gamma rays which are emitted when the positrons strike a metal collector plate at the end of the RPA. Both the collision cell and the RPA are located in a magnetic field of $\sim$0.1 T.

In order to measure scattering cross sections in the presence of the magnetic field, we exploit the properties of the positron orbits in the field. The total energy of a positron, E_{T}, can be separated into two components

$$E_{\mathrm{T}} = E_{\parallel} + E_{\perp}$$

where $E_{\parallel}$ is the energy in the motion parallel to the field and $E_{\perp}$ is the energy in the cyclotron motion perpendicular to the field. In the present experiments an incident beam with $E_{\parallel} > 8$ eV and $E_{\perp} \sim$0.025 eV is used. The RPA only measures $E_{\parallel}$. Many scattering processes can change $E_{\parallel}$ and, at the energies considered here, both elastic and inelastic scattering (i.e. rovibrational and electronic) are possible. For each of these processes, the positron will also be scattered through some scattering angle θ. As a result, these scattering events can transfer some of the total energy from $E_{\parallel}$ into $E_{\perp}$. Denoting the initial positron energy as E_{i} and the total energy of the positron after scattering as E_{s} then

$$E_{\mathrm{s}} = E_{\mathrm{i}} \quad \text{(elastic scattering)} \tag{1}$$

and

$$E_{\mathrm{s}} = E_{\mathrm{i}} - E_{\mathrm{ex}} \quad \text{(inelastic scattering).} \tag{2}$$

For both elastic and inelastic scattering,

$$E_{\parallel} = E_{\mathrm{s}} \cos^2(\theta), \tag{3}$$

and

$$E_{\perp} = E_{\mathrm{s}} \sin^2(\theta), \tag{4}$$

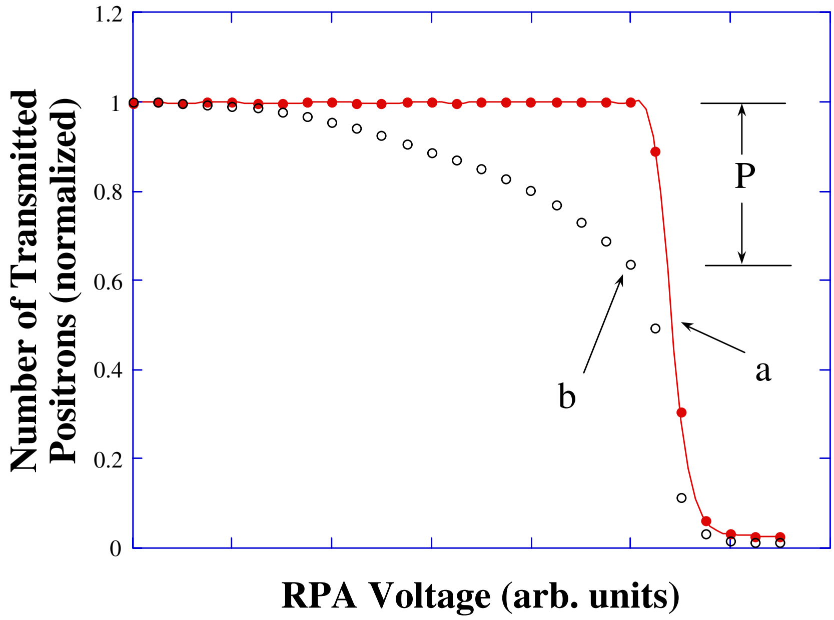

Figure 1. Example of an RPA cut-off curve measured with (○) and without (●) gas in the scattering cell. The beam strength has been normalized to unity. The points labelled are a, the beam cut-off, and b, the point at which the total cross section measurement is made. P is the integrated probability of the occurrence of any scattering event. See text for details.

where E_{ex} is the energy loss involved in an excitation process. Thus all scattering processes (except forward and backward elastic scattering) will lead to a loss of $E_\parallel$, and this will be reflected in the signal strength measured by the RPA.

This is demonstrated in figure 1, which illustrates typical RPA cut-off curves obtained with and without gas in the scattering cell. Without gas in the cell, positrons are transmitted uniformly right up to the cut-off point, labelled by a, where the retarding potential equals E_i/e. The width of the cut-off is a measure of the energy resolution of the beam. With gas in the cell, the effects of scattering can be clearly seen at voltages below the cut-off point. In this region, positrons have lost $E_\parallel$ due both to angular scattering in elastic and inelastic collisions, and they are cut-off at lower RPA voltages. From equations (1)–(4), and figure 1, it can be seen that this collisional loss of $E_\parallel$ extends all the way back to zero energy as, for example, would occur in an elastic collision at $\theta = 90°$ for which $E_\parallel = 0$ and $E_\perp = E_i$. If $\theta > 90°$, the positrons are scattered back toward the trap; but they are then reflected from the potential wall that defines the end of the trap and they pass through the gas cell once more. Thus, under the assumption that they undergo no further collisions, it is impossible to distinguish between scattering through $\theta°$ and $(180 - \theta)°$. For the present measurements of the grand total cross section, this lack of angular discrimination is of no significant consequence.

As the RPA curve represents an integral spectrum, if we monitor the transmitted positron signal at a voltage close to the beam energy cut-off (e.g. the point labelled by b in figure 1), then the difference between this signal level and the unscattered beam strength (normalized to unity) is the probability of any scattering event occurring. This is denoted by P in figure 1 and it is related to the total scattering cross section (Q_t) by

$$Q_t = \frac{P}{nl}, \qquad (5)$$

Table 1. Summary of resonance searches.

Target	Energy range (eV)	Possible parent states	Upper bound on resonance strength (a_0^2 meV)
H_2	10.4–12.8	B $^1\Sigma$, C $^1\Pi$, E $^1\Sigma$, F $^1\Sigma$	2.0
N_2	7.9–8.9	a′ $^1\Sigma$, a $^1\Pi$, w $^1\Delta$	2.0
CO	5.0–10.0	A $^1\Pi$, I $^1\Sigma$, D $^1\Delta$	11
Ar	11.0–12.0	$3p^5(^2P_{3/2,1/2})\,4s\ (J = 1)$	6.3

where n is the gas number density and l the length over which the scattering takes place (Gilbert *et al* 2000). In the present experiments, l is taken to be the physical length of the scattering cell (38.1 cm), and n corresponds to the number density of the target gas in the cell. For figure 1, the value of $P \sim 0.4$ was chosen to illustrate the technique. For the scattering measurements P was typically kept to less than 0.1.

To ensure that the spread in beam energy does not affect the measurement, the voltage (b) is chosen to be three standard deviations ($\sim$40 meV) from the beam cut-off. For the collision energies studied here, this corresponds to an elastic scattering angle of $\leqslant 4°$ (and $\geqslant 176°$) and thus, although the majority of the angular range is accounted for, the present measurements are a slight underestimate of the total cross section. However, given the $\sin\theta$ weighting of the differential scattering cross section in the total cross section, this is a small effect ($\sim$1%) for the cross sections measured here. We also note that the present grand total cross section measurements include the effects of positronium formation at energies where that channel is open.

In estimating the effective sensitivity of our measurements to the possible presence of a resonance, we have taken the following approach. We denote the resonance cross section and width by σ_r and ΔE_r, the limiting uncertainty on the measurements as $2\sigma_{rms}$, where σ_{rms} is the statistical uncertainty (one standard deviation) on the data, the beam energy width by ΔE_b and the energy step size in the measurements by ΔE_m. In the present case we have assumed that $\Delta E_r < \Delta E_b$, ΔE_m is most likely for all targets and, as a result, the limiting resonance strength, $\sigma_r \Delta E_r$, is given by (95% confidence level)

$$\sigma_r \Delta E_r < 2\sigma_{rms} \Delta E_b. \qquad (6)$$

For $\Delta E_r > \Delta E_b$, ΔE_m the limit placed on the resonance strength would be increased modestly.

The absolute energy scale was established by calibrating the potential difference between the positron source and the scattering cell against the known threshold energies for processes such as vibrational excitation. The estimated uncertainty in the beam energy is $\sim$25 meV.

3. Results and discussion

We have measured absolute total cross sections for positron scattering from H_2, N_2, CO and Ar at incident energies which range from a few eV below the first electronic excited state threshold to a few eV above. For all of these measurements, the energy spread in the incident beam was $\sim$25 meV and cross section measurements were made at energy intervals of 25 meV for H_2, N_2 and Ar, and 50 meV for CO. In the case of H_2, some of the data were also taken with an energy interval of 10 meV. The measurements are summarized in table 1.

Figure 2 shows the total cross section for H_2 at energies between 10.4 and 12.8 eV. This covers an energy range which extends from about 0.8 eV below the threshold for the B $^1\Sigma$

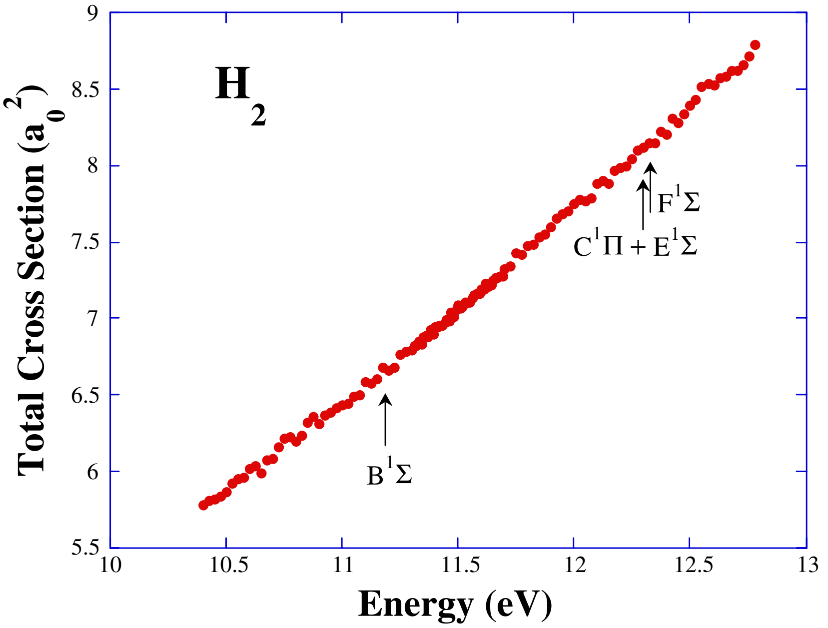

Figure 2. The total cross section for positron scattering from H_2. The arrows mark the energies of the thresholds for specific electronic excited states.

state to about 0.5 eV above the threshold for excitation of the F $^1\Sigma$ state. In particular it encompasses the energy region where Varella *et al* have predicted the existence of a Feshbach resonance. While the absolute value of the cross section is of importance in comparison with other data, this will be the subject of a future communication. Of paramount importance in the present study has been the achievement of high statistical accuracy. In figure 2 the statistical uncertainty (one standard deviation) of the measured points is $\sim$0.04 a_0^2, which is smaller than the point size on the plot. It is clear from figure 2 that we do not see any feature of the prominence predicted by Varella *et al* (2001). The 'strength' of their resonance feature in the integral elastic cross section was predicted to be about 100 a_0^2 meV. From the present measurements of the total cross section, we can conservatively estimate, based on two standard deviations, an upper limit on the strength of the resonance to be 2 a_0^2 meV. This is a factor of 50 smaller than the predicted value. In the present case we expect that there may be some loss of sensitivity in the measurement as we are measuring the grand total scattering cross section, whereas the prediction was for the integral elastic cross section. However, in the energy region of the predicted resonance, below the first electronic excitation threshold, the only open inelastic channels are rovibrational excitation and positronium formation. The former are expected to contribute (at most) a few per cent to the total cross section at these energies, whilst the latter, with a threshold at 8.63 eV, may be responsible for as much as 20% of the total scattering (Kwan *et al* 1998). Thus we estimate that the resonance, if it exists, is substantially weaker than the calculated structure of Varella *et al*.

In figure 3 we show total cross section measurements for CO in the energy region between 5 and 10 eV, which encompasses the threshold for positronium formation (7.2 eV) and the excitation thresholds for the I $^1\Sigma$ (8.003 eV), A $^1\Pi$ (8.027 eV) and D $^1\Delta$ (8.108 eV) states. The effect of positronium formation is clearly evident above 7.2 eV; however, there is no evidence for any sharp, resonance-like features in the cross section. The upper limit that we

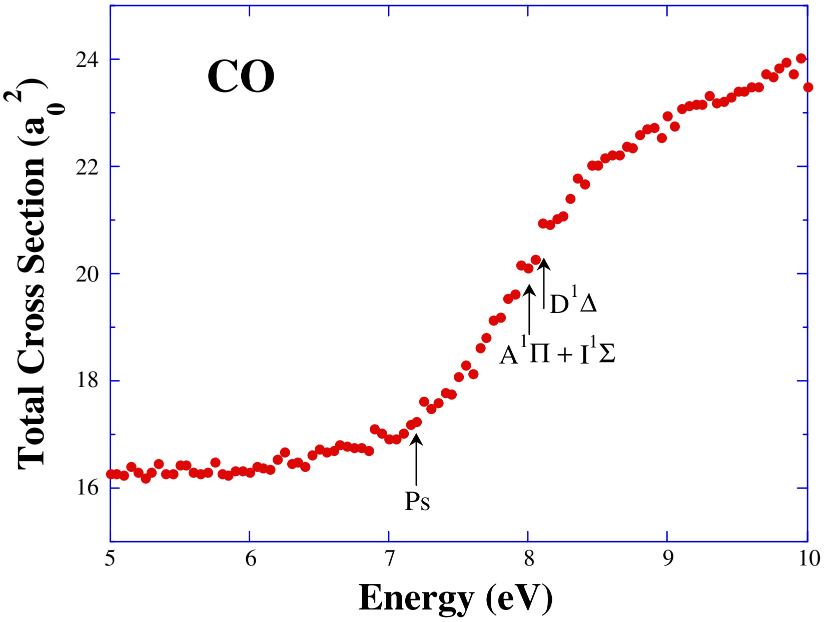

Figure 3. The total cross section for positron scattering from CO. The arrows mark the energies of the thresholds of a number of electronic excited states and positronium (Ps) formation.

can place on the strength of any possible resonance is larger ($\sim$11 a_0^2 meV) as the statistical accuracy of the data is not as high as for H_2. What may appear to be a small bump in the cross section between 8 and 9 eV could be due to the onset of electronic excitation. Recent measurements (Sullivan *et al* 2001b) have shown electronic excitation of these states to be quite strong at near-threshold energies.

The results for N_2 and Ar are similar to those for H_2 and CO. For N_2, no obvious resonance features were observed (limiting strength of $\sim$2 a_0^2 meV) between 7.9 and 8.9 eV, the region of the excitation thresholds of the a$'\,^1\Sigma$, a $^1\Pi$ and w $^1\Delta$ states. The thresholds for these electronic excited states also lie below the threshold for positronium formation in N_2 (8.78 eV) and so elastic scattering is the main contributor to the total cross section. In argon, the total cross section was measured between 11 and 12 eV, encompassing the excitation thresholds for the 3p^5 ($^2P_{3/2}$) 4s ($J = 1$) and 3p^5 ($^2P_{1/2}$) 4s ($J = 1$) states, which lie at 11.63 and 11.82 eV respectively. Once again there was no evidence of any strong resonance feature with an upper bound on the strength of 6.3 a_0^2 meV. All of the above measurements and the resonance strength limits are summarized in table 1.

In conclusion we have made precise measurements of the grand total positron scattering cross sections for H_2, N_2, CO and Ar with the goal of searching for Feshbach resonances associated with the lowest electronically excited states of these targets. No prominent resonance features were observed in any of these targets. Based on these measurements, we believe that a conservative upper limit on the strength of any such resonances in the energy regions studied is in the range 2–11 a_0^2 meV, depending on the target. In particular the results for H_2 do not confirm a recent Schwinger variational calculation (Varella *et al* 2001), which predicts a strong Feshbach resonance in the elastic scattering channel below the threshold for the excitation of the B $^1\Sigma$ state. While the total cross section is not the most sensitive scattering channel to search for narrow Feshbach resonances, such features have been observed, often with greater strengths than the present level of sensitivity, for electron scattering from a variety of atoms and some simple molecules. In the case of positron scattering it may be that more sensitive, state-specific integral cross section measurements or angular differential cross section measurements are required to reveal what are most likely to be (at best) weak scattering processes. Such studies, particularly those involving integral cross section measurements, are also possible with the cold positron beam described here. We hope that this letter, describing new tools for the study of positron resonances, or quasi-bound states, may provide stimulation for further extension and refinement of the theoretical calculations on positron resonances.

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References

Buckman S J and Clark C W 1994 *Rev. Mod. Phys.* **66** 539
Campbell C P, McAlinden M T, Kernoghan A A and Walters H R J 1998 *Nucl. Instrum. Methods* B **143** 41
Gien T T 1996 *Can. J. Phys.* **74** 343
Gilbert S J, Kurz C, Greaves R G and Surko C M 1997 *Appl. Phys. Lett.* **70** 1944
Gilbert S J, Greaves R G and Surko C M 1999 *Phys. Rev. Lett.* **82** 5032
Gilbert S J, Sullivan J P, Greaves R G and Surko C M 2000 *Nucl. Instrum. Methods* B **171** 81
Gribakin G F 2000 *Phys. Rev.* A **61** 022720
Iwata K, Gribakin G F, Greaves R G, Kurz C and Surko C M 2000 *Phys. Rev.* A **61** 022719
Kwan C K, Kauppila W E, Nazaran S, Przybyla D, Scahill N and Stein T S 1998 *Nucl. Instrum. Methods* B **143** 61
Mitroy J, Bromley M W J and Ryzhikh G 1999 *J. Phys. B: At. Mol. Opt. Phys.* **32** 2203
Mittleman M H 1966 *Phys. Rev.* **152** 76
Ryzhikh G G and Mitroy J 1997 *Phys. Rev. Lett.* **79** 4124
Smith P M and Paul D A L 1970 *Can. J. Phys.* **48** 2984
Stein T S, Laperriere F, Dababneh M S, Hsieh Y-F, Pol V and Kauppila W E 1981 *12th Int. Conf. on the Physics of Electronic and Atomic Collisions (Gatlinburg, TN)* ed S Datz, contributed abstracts p 424
Sullivan J P, Gilbert S J and Surko C M 2001a *Phys. Rev. Lett.* **86** 1494
Sullivan J P, Gilbert S J, Buckman S J and Surko C M 2001b *Bull. Am. Phys. Soc.* **46** 58
Surko C M, Passner A, Leventhal M and Wysocki F J 1988 *Phys. Rev. Lett.* **61** 1831
Varella M T N, Carvalho C R C and Lima M A P 2001 *New Directions in Antimatter Chemistry and Physics* ed C M Surko and F A Gianturco (Amsterdam: Kluwer) p 493