

LETTER TO THE EDITOR

Analysis of $1s2l2l'$ and $2l2l'2l''$ resonances of He^- ions

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Abstract. Hyperspherical adiabatic potential curves for the three-electron He^- ions are calculated. Potential curves that support $1s2l2l'$ doubly excited states and $2l2l'2l''$ triply excited states are analysed to interpret the possible existence and the nature, including the relative autoionization widths, of these states. We show that doubly excited states of He^- can be classified using the same set of quantum numbers K , T and A that were used for classifying doubly excited states of two-electron atoms. We also show the relative energy levels of triply excited states exhibit a rotor structure.

An electron cannot be attached to the ground state of a helium atom to form a stable helium negative ion. However, it is well known that there exist He^- resonances associated with the singly and doubly excited states of He (see Buckman and Clark 1994). In this letter we will address the doubly excited $1s2l2l'$ resonances and the triply excited $2l2l'2l''$ resonances of He^- . The first group of resonances lie at about 19–22 eV and the second group lie at about 57–60 eV, respectively, above the ground state of He. There are other He^- doubly and triply excited states associated with other thresholds, but only these two groups will be addressed in this letter.

Experimentally resonances in He^- have been studied repeatedly over the last four decades. The challenge is to improve electron energy resolution such that the detailed structure in the spectra can be analysed with confidence. Many spurious resonances have been ‘discovered’ over the years and then discarded in later experiments. The reader can find a more complete account of this history in the review article by Buckman and Clark (1994). On the theoretical side there have been a number of elaborate calculations on electron–helium atom collisions. Elastic or inelastic scattering cross sections have been calculated. Both experiments and theories attempted to locate and identify resonances from the structures of the scattering cross sections. However, many such structures occur near the inelastic thresholds and cannot be easily disentangled from the cusps which often accompany the opening of new inelastic channels.

A powerful tool for the analysis of resonances is the hyperspherical method within the adiabatic approximation. In this approach, bound and resonance states are not calculated directly. Instead, the hyperspherical adiabatic potential curves are first calculated. If a potential curve has a relatively deep well, then it is likely that some bound or resonance states can be found. If the potential curve is fully repulsive, then it cannot support bound states. This method has been fully developed for the two-electron atoms (Lin 1986, 1993) and other three-body systems such as the helium trimer (Esry *et al* 1996), as well as the positron–hydrogen collision system (Zhou and Lin 1995). Beyond the adiabatic approximation,

the adiabatic expansion of the total wavefunctions has been used to obtain state-of-the-art quantitative results for various systems (Tang *et al* 1992a, Tolstikhin *et al* 1996, Kato and Watanabe 1997, Chen *et al* 1997). Photoabsorption cross sections of H^- and He (Tang *et al* 1992b, 1994) have been calculated and shown to agree with the most detailed experiments done so far.

The hyperspherical approach has been extended to three-electron atoms. Earlier works, in general, do not provide enough accuracy. For the three-electron He^- ion, quantitative results for doubly excited states have been obtained by Watanabe and co-workers (Watanabe 1982, Le Dournerf and Watanabe 1990) earlier. They treated the doubly excited states of He^- as a two-electron system where the two outer electrons move in a frozen open-shell $\text{He}^+(1s)$ core. The potential curves for the two-electron systems were calculated and used to analyse the doubly excited states of He^- ions. Many insights have emerged from this approach. In two conference reports (Watanabe *et al* 1982, Watanabe 1988), Watanabe also presented quantitative calculations for some triply excited states where the potential curves were calculated by diagonalizing the Hamiltonian with basis functions constructed from products of Slater-type orbitals.

In this letter we will report some results on He^- ions calculated using the *three-electron* hyperspherical approach. This allows us to study all the states in a three-electron system, including excitations of one, two or three electrons, all together at the same time. We will

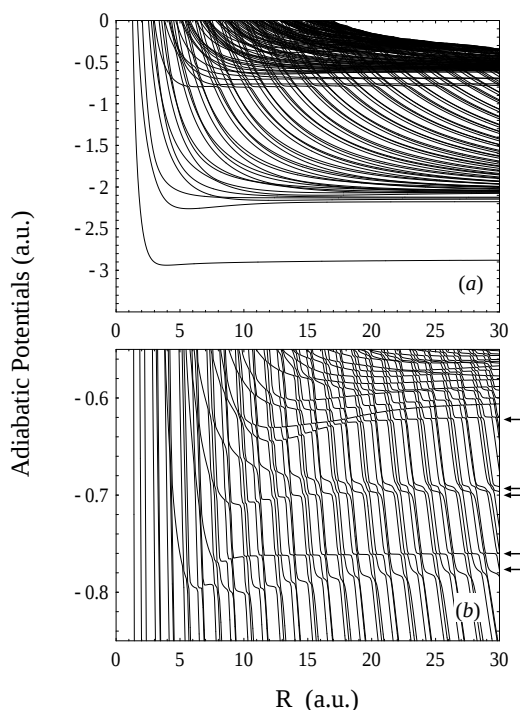


Figure 1. (a) Hyperspherical potential curves for the $2S^e$ symmetry of the He^- ion. (b) An expanded view of the higher-energy region showing potential curves that support triply excited states have numerous avoided crossings with the steep curves that support doubly excited states. In the ‘adiabatic’ approximation these avoided crossings will be treated as diabatic. The lowest five curves for the triply excited states converge to the $2s^2\ ^1S^e$, $2p^2\ ^3P^e$, $2p^2\ ^1D^e$, $2s2p\ ^1P^o$ and $2p^2\ ^1S^e$ states of He, each marked by an arrow on the right.

focus on the qualitative aspect of these doubly and triply excited resonances as revealed from the analysis of potential curves. More accurate quantitative calculations for individual states require future careful treatment of the couplings of the adiabatic channels.

The detailed hyperspherical calculations for three-electron atoms or ions have been presented elsewhere (Morishita and Lin 1997, Morishita *et al* 1998). For the purpose of this letter, we mention that within the adiabatic approximation where the hyperradius is treated as an adiabatic parameter, a family of potential curves can be obtained. For example, in figure 1(a) we show the family of potential curves for the $^2S^e$ states of He^- . The lowest curve is repulsive (an enlarged scale would show this fact more clearly), indicating that an electron cannot attach to the ground state of He to form a stable He^- bound state. The next group of curves converge to the singly excited $\text{He}(1snl)$ states asymptotically as $R \rightarrow \infty$. These potential curves support doubly excited states of He^- . Further up on the energy scale, we can identify the family of curves which converge to the doubly excited states of He. These curves support triply excited states and they have numerous avoided crossings with the curves that support doubly excited states. The curves that converge to the $\text{He}(2l2l')$ doubly excited states are displayed in figure 1(b) to show the details.

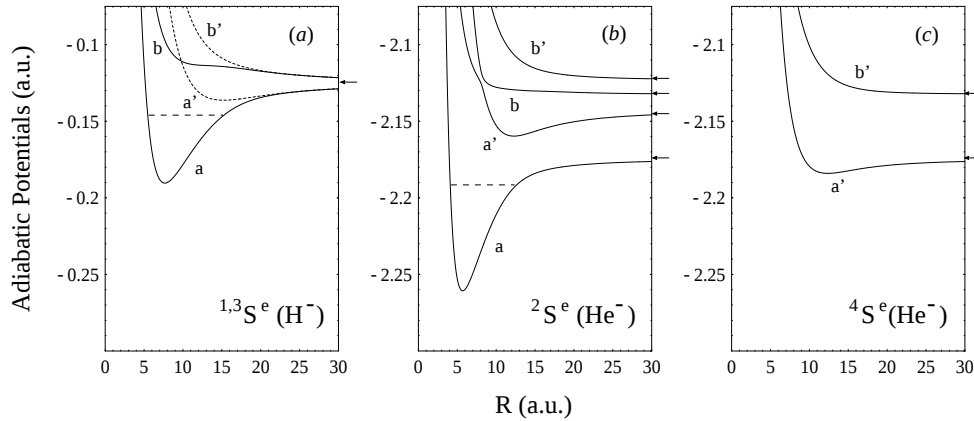


Figure 2. Comparison of the $1,3S^e$ potential curves of H^- with the $2S^e$ and $4S^e$ curves of He^- . For H^- these curves support $2lnl'$ doubly excited states. For He^- these curves support $1s2lnl'$ doubly excited states. The symbols that label the curves are explained in the text. We used arrows to indicate the asymptotic limits of the potential curves. For H^- , the arrow indicates the energy of $\text{H}(n=2)$. For He^- , the four arrows indicate the energies of $1s2s\ ^3S^e$, $1s2s\ ^1S^e$, $1s2p\ ^3P^o$ and $1s2p\ ^1P^o$ of He, respectively, from the bottom. The horizontal broken curves indicate the positions of the known resonances.

We first focus on the doubly excited states. The four curves that converge to $\text{He}(1s2l)$ states are shown in figure 2(b). They are compared to the $1S^e$ and $3S^e$ potential curves of H^- . We have used 'a' and 'b' as short-hand notation to label the two $1S^e$ potential curves of H^- in figure 2(a). In terms of the K , T and A quantum numbers for the doubly excited states of two-electron atoms (Herrick and Sinanoğlu 1975, Lin 1984, 1986), $a = (1, 0)^+$ and $b = (-1, 0)^+$ for the $1S^e$ curves. Similarly, we use $a' = (1, 0)^-$ and $b' = (-1, 0)^-$ to label the two $3S^e$ curves. These symbols are also used to label the He^- potential curves in figures 2(b) and (c). Clearly, the two sets of curves in figures 2(a) and (b) are quite similar in the inner region. They differ in the asymptotic region since the curves in figure 2(a) all converge to the degenerate $\text{H}(n=2)$ limit and for figure 2(b) they converge to the four different limits, namely, the $1s2s\ ^{1,3}S^e$ and $1s2p\ ^{1,3}P^o$ states of He.

The similarity of the curves in figure 2(b) to those in figure 2(a) for H^- has been elaborated by Watanabe (1982). In the range of $R = 6-10$, which is near the minima of the curves, the He^- potential describes the situation of a tightly bound 1s electron and two outer ones which are nearly at the same distance from the nucleus. The 1s electron fully screens the He^{2+} nucleus such that the two electrons experience only one unit of positive charge, similar to that for the H^- ion. Therefore the He^- potential curves in this region are similar to those for H^- . At large R , we consider the states such as $1s2lnl'$ ($n \gg 2$) states, where the outermost nl' electron couples weakly to the other two electrons. The coupling between the 1s and $2l$ electrons is stronger and the potential energy curves are designated by the $\text{He}(1s2l)$ singly excited states at large R .

For H^- there is only one intra-shell doubly excited state among the curves shown in figure 2(a). It is the $2s^2\ ^1\text{S}^e$ state (for simplicity we use the conventional independent particle notation to describe the doubly excited states; in general, doubly excited states are better designated with the K , T and A quantum numbers), which is supported by curve a. Curve a' also supports some Feshbach resonances that are very near the threshold, but these inter-shell resonances are specific to the true two-electron systems only and they are bound by the attractive dipole potential (goes like R^{-2} at large R). For He^- , curve a in figure 2(b) is deep enough to support a resonance, designated as $1s2s^2\ ^2\text{S}^e$ if we are allowed to use the independent particle description. The curve a' for He^- has a shallow attractive well, but it is not deep enough to support a resonance. Unlike H^- , there is no attractive dipole potential for the $e^- + \text{He}(1s2l)$ system asymptotically and no resonances can be supported by the dipole polarization potential which goes like R^{-4} at large R . In figure 2(b), curves b and b', like their counterparts in H^- , are completely repulsive and cannot support any resonances. We have used horizontal broken lines to indicate the positions of the known resonances for each ion.

In figure 2(c) we show the two $^4\text{S}^e$ curves of He^- . They are similar to the two $^3\text{S}^e$ curves in H^- at small R , but converge to the $1s2s\ ^3\text{S}^e$ state and $1s2p\ ^3\text{P}^o$ state, respectively, at large R . They are labelled by a' and b' and both curves are not attractive enough to support bound or resonance states.

Before discussing the higher curves in figure 1, we examine another example of doubly excited states of He^- . In figures 3(a)–(c), we show the curves for $\text{H}^-(^1,^3\text{P}^o)$, $\text{He}^-(^2\text{P}^o)$ and $\text{He}^-(^4\text{P}^o)$, respectively. For H^- , a = $(1, 0)^+\ ^3\text{P}^o$ and a' = $(0, 1)^+\ ^1\text{P}^o$. The other curves are not important for the resonances and will not be labelled or discussed. In H^- , curve a supports the $2s2p\ ^3\text{P}^o$ Feshbach resonance and a' (which is allowed to cross the next upper curve at about $R = 12$) supports the $2s2p\ ^1\text{P}^o$ shape resonance (Lin 1975). In $\text{He}^-(^4\text{P}^o)$, the 1s electron is coupled to the outer pair of electrons in the $2s2p\ ^3\text{P}^o$ state such that a doubly excited state, designated as $1s(2s2p\ ^3\text{P}^o)^4\text{P}^o$, can be formed. The potential curve that supports this state is essentially the same as the one that supports the $2s2p\ ^3\text{P}^o$ state of H^- . In fact, by comparing figures 3(a) and (c), we note that the two potential curves labelled as 'a' are quantitatively almost identical if one subtracts the 1s energy of the He^+ (−2 au) from the actual values of curve a in figure 2(c). For $^2\text{P}^o$ states, the situation is more complicated since there is a clearly noticeable avoided crossing between curves a and a' at about $R = 10-12$. If the crossing is treated adiabatically then each potential alone is not strong enough to support a resonance. However, this crossing has to be treated carefully, at least in a two-channel approximation including curves a and a'. In fact, as shown by Watanabe (1982), the experimentally observed structure for the $^2\text{P}^o$ symmetry at 20.4 eV, which is above the $\text{He}(1s2s\ ^3\text{S}^e)$ threshold but below the $\text{He}(1s2s\ ^1\text{S}^e)$ threshold, cannot be assigned to a particular channel (or a potential curve). In fact, in the single-channel approximation it is a better approximation to treat this avoided crossing as a

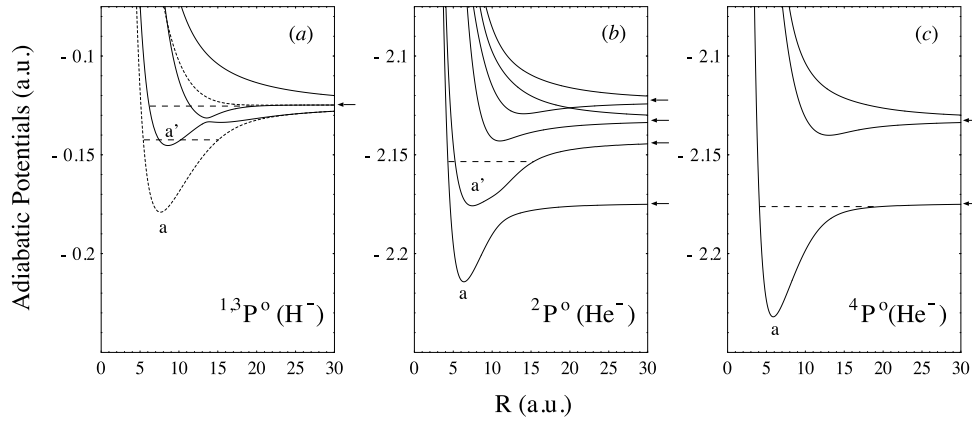


Figure 3. Comparison of the $1,3P^o$ potential curves of H^- with the $2P^o$ and $4P^o$ curves of He^- . For H^- these curves support $2nl'$ doubly excited states. For He^- these curves support $1s2nl'$ doubly excited states. For H^- the avoided crossing for curve a' near $R = 12$ with the next upper curve is to be treated diabatically. For the $2P^o$ symmetry of He^- , the pronounced avoided crossing between a and a' near $R = 12$ implies that the coupling between these two channels is large. In a rough single-channel approximation this avoided crossing is to be treated diabatically, but the coupling implies a large autoionization width for the $1s2s2p\ 2P^o$ resonance. For H^- , the arrow indicates the energy of $H(n = 2)$. For He^- , the four arrows indicate the energies of $1s2s\ ^3S^e$, $1s2s\ ^1S^e$, $1s2p\ ^3P^o$ and $1s2p\ ^1P^o$ of He, respectively, from the bottom. The horizontal broken lines indicate the positions of the known resonances.

diabatic crossing, then the structure at 20.4 eV may be viewed approximately as a Feshbach resonance associated with a ‘diabatic’ channel which coincides with curve a at small R before the crossing and with curve a' at large R after the crossing. The coupling of these two channels results in a large width for this state. From various calculations the width of this structure is about 400 meV (Buckman and Clark 1994) which is much larger than the width of, say, the $1s2s^2\ ^2S^e$ resonance, which is about 11 meV.

For completeness, we mention that there are two other intra-shell resonances in H^- , one is the $2p^2\ ^3P^e$ state and the other is the $2p^2\ ^1D^e$ state. For He^- laser spectroscopy (Walter *et al* 1994) from the metastable state $1s2s2p\ ^4P^o$ of He^- clearly indicates that there is a $4P^e$ shape resonance which can be viewed as the $1s(2p^2\ ^3P^e)^4P^e$ state, i.e. from coupling the $1s$ to the $2p^2\ ^3P^e$ state of the electron pair. We comment that for H^- the $2p^2\ ^3P^e$ state is about 10 meV below the $H(n = 2)$ threshold, but the $1s(2p^2\ ^3P^e)^4P^e$ state is 12.3 meV (Walter *et al* 1994) above the $1s2p\ ^3P^o$ threshold of He. At even higher energies, there is another structure for the $2D^e$ symmetry which is very near the $He(1s2p\ ^3P^o)$ threshold. This structure had been attributed to be a $1s(2p^2\ ^1D^e)^2D^e$ resonance. However, as shown by Watanabe (1982), there are no hyperspherical adiabatic potential curves to support such a resonance state. The $2D^e$ feature in the experimental data is the combined effect of threshold behaviour and the coupling of the relevant potential curves. The reader is advised to consult the paper by Watanabe (1982) for further details on this ‘resonance’.

Thus among the doublet states in the 19–21.5 eV region only the $2S^e$ state is a clear-cut Feshbach resonance, the $2P^o$ and $2D^e$ resonances or structures are less clear cut; they cannot be viewed as associated with a single channel, at least in the hyperspherical sense. For the quartet states, the known resonances are the $1s2s2p\ ^4P^o$ and $1s2p^2\ ^4P^e$ states. Each of them can be identified as the bound state or shape resonance associated with a single hyperspherical potential curve. Beyond these intra-shell resonances (or structures)

discussed above, there are no other inter-shell doubly excited resonances associated with $\text{He}(1s2l)$ thresholds.

We proceed next to discuss the $2l2l'2l''$ triply excited states of He^- . For $^2S^e$, from figure 1(b) it is clear that the potential curves that support triply excited states have numerous avoided crossings with curves that support doubly excited states. We treat each of these crossings diabatically such that potential curves associated with triply excited states for each symmetry can be extracted. In this letter we focus on intra-shell triply excited states. Among the potential curves that converge to the $2l2l'$ doubly excited states of He, only those curves that are relevant to the $2l2l'2l''$ triply excited states will be considered. The identification of such curves is, in general, simple. These curves have deep attractive wells at small values of R . However, in certain cases, there is evidence of strong avoided crossings among the curves that support triply excited states. The two lowest $^2S^e$ curves that possibly can support triply excited states (see figure 1(b)) have a pronounced avoided crossing. (This avoided crossing is even more clear in the case of the isoelectronic Li.) Within the single-channel approximation we drew the curves diabatically, such that the lowest $^2S^e$ curve converges to the $2s2p\ ^3P^o$ limit of He asymptotically instead of the lower $2s^2\ ^1S^e$ limit. (This single-channel approximation is chosen for the moment for its simplicity. We expect that at least a two-channel approximation is needed to account for the coupling of these two lowest channels.)

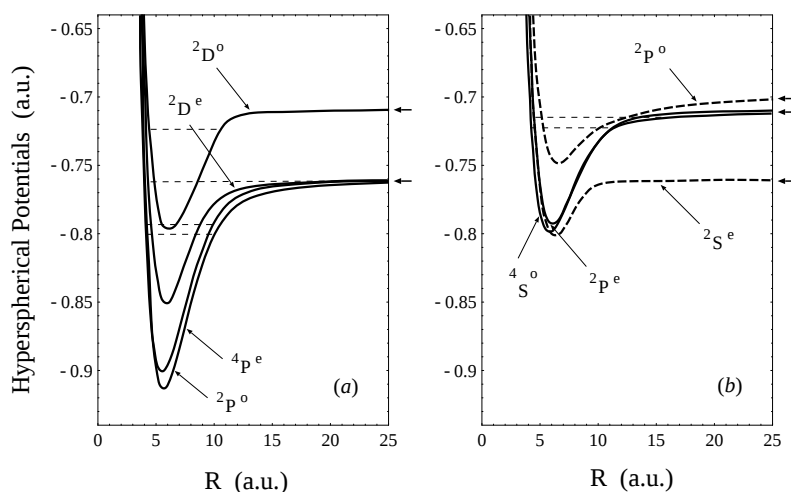


Figure 4. Comparison of potential curves that can possibly support $2l2l'2l''$ intra-shell triply excited states of He^- . The group of curves in (a) are related approximately as the rotational excitations of a symmetric top formed by the three electrons. The curves show that the $^2S^e$ curve and the second $^2P^o$ curve are not deep enough to support resonance states. All the other six symmetries can support intra-shell triply excited states. The horizontal broken lines indicate the positions of the known resonances.

We have calculated all the potential curves for the total angular momentum $L = 0, 1$ and 2 for He^- . In figure 4 we show the eight potential curves that have relatively deep wells which have the possibility of supporting $2l2l'2l''$ intra-shell triply excited states. These states can be seen as resonances in electron–helium collisions (e.g. Kuyatt *et al* 1965, also see Buckman and Clark 1994) and in beam-foil experiments (e.g. Knystautas 1992). They also have been studied by various theoretical methods since the first work of Fano and Cooper (1965). Most recently, Bylicki and Nicolaides (1995) have used the complex coordinate

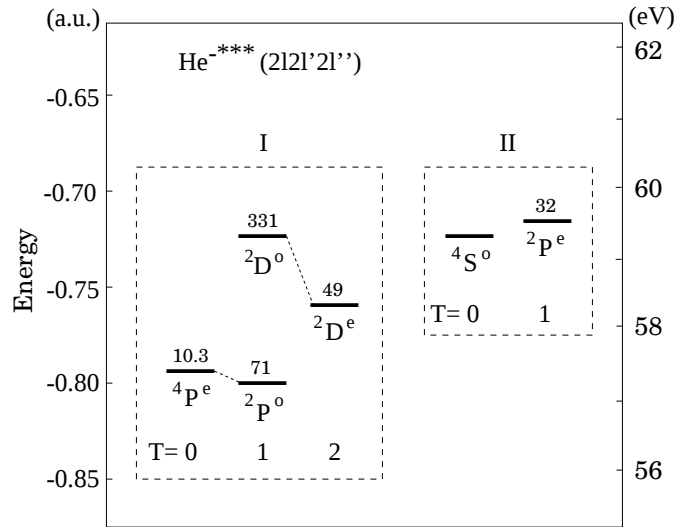


Figure 5. Energies and autoionization widths of intra-shell $2l2l'2l''$ triply excited states of He^- . The numbers above each state give the resonance width in meV. These data are taken from the calculations of Bylicki and Nicolaides (1995) and Davis and Chung (1990). The four states in the first group (I) form a (truncated) rotor series similar to that from a symmetric top. These states have the shape of an equilateral triangle with the nucleus at the centre, i.e. it is coplanar. The three electrons for the two states in group II also have the most probable shape of an equilateral triangle, but the wavefunction vanishes when the plane contains the nucleus, i.e. the coplanar geometry is forbidden.

rotation method to calculate the positions and widths of the possible $2l2l'2l''$ resonances. Their results are displayed in figure 5. The nonautoionizing $4S^o$ state calculated by Davis and Chung (1990) is also included. The resonance energies are given with respect to the ground state of He.

We wish to ‘interpret’ the results in figure 5 in terms of the potential curves shown in figure 4. We comment that the energies calculated using a one-channel approximation from the curves in figure 4 will not be as accurate as those shown in figure 5, except possibly after the coupling with other channels is included.

First, the four curves shown in figure 4(a) all support resonances. In fact, from the analysis of the nodal structure (Bao *et al* 1997, Watanabe and Lin 1987) of intra-shell state wavefunctions, the energies of intra-shell states associated with these four curves are approximated by

$$E(L, T) = \frac{1}{2I} [2L(L+1) - T^2] + E_0 \quad (1)$$

where the quantization axis of the body frame is perpendicular to the plane formed by the three electrons, T is the projection of L along this axis, I is the moment of inertia and E_0 is the energy term associated with the internal motion. I and E_0 are approximately independent on the four states. The T ’s for the four states, counting from the bottom curve, are 1, 0, 2 and 1, respectively. For the $2P^o$, $4P^e$ and $2D^e$ curves, each curve is the lowest one that supports triply excited states and there are no obvious avoided crossings with other curves of that symmetry. The $2D^o$ potential curve is somewhat different. The lowest curve has a pronounced avoided crossing with the next curve. The two lowest curves are similar to curves a and a’ in figure 3(b). We drew these curves diabatically in

the present single-channel approximation such that the $^2D^o$ curve converges to the higher threshold. The avoided crossing, similar to the case in figure 3(b), implies that this state would decay readily and the calculation of Bylicki and Nicolaides (1995) gives this state a width of 331 meV, which is much larger than the widths of 10–70 meV for the other three states. We also point out that the four potential curves in figure 4(a) display patterns that are characteristic of the rotation of a symmetric top. From the analysis of Bao *et al* (1997) and Watanabe and Lin (1987), the three electrons in these states form an equilateral triangle with the nucleus at the centre. The energy differences of these states are due to the rotational excitations.

We next consider the four other curves in figure 4(b). The curves for $^4S^o$ and $^2P^e$ are, respectively, the lowest curve of each symmetry and there are no avoided crossings with other curves. Each curve is deep enough to support a resonance state. For the $^2S^e$ curve, as seen from figure 1(b), the two lowest curves have a pronounced avoided crossing. In a one-channel approximation we treat this crossing diabatically. However, even under this approximation each of the resulting curves is not attractive enough to support a resonance. We thus conclude that there are no $^2S^e$ triply excited states in He^- , in agreement with the conclusion of Bylicki and Nicolaides (1995). The only remaining possible intra-shell $2/2'/2''$ triply excited state would be the $2p^3\ ^2P^o$ state. We have extracted the potential curve within the single-channel approximation and the resulting curve is shown in figure 4(b). Its potential well is not deep enough to support a resonance. This conclusion again is in agreement with Bylicki and Nicolaides (1995). We comment that the potential curves shown in figure 4 clearly explain the relative order of the energies of the $2/2'/2''$ states of He^- , see figure 5. Since the $^4S^o$ curve shown in figure 4(b) is the lowest adiabatic potential curve (there are no singly or doubly excited $^4S^o$ states), we can calculate the energy of this state within the single-channel approximation including the second-order diagonal coupling term. The value we obtained is -0.7201 au, which is in good agreement with the value $-0.722\,952$ au obtained by Davis and Chung (1990).

In summary we have calculated the hyperspherical adiabatic potential curves for the He^- ion. A subset of these potential curves were used to analyse the $1s2/2'$ doubly excited states and the $2/2'/2''$ triply excited states. We found that most of these states can be described as bound states of individual single adiabatic potential curves. When resonances do not exist for a given symmetry, the corresponding potential curves are, in general, repulsive, i.e. there is no potential well or the potential well is not deep enough to support a bound or resonance state. In a number of cases the single-channel approximation fails. We found cases where two potential curves show pronounced avoided crossings. In this case the resonance widths, in general, are very broad. We pointed out that the $^2P^o$ doubly excited state and the $^2D^o$ triply excited state both belong to this case and each state has a very large autoionization width. We have also shown that the $1s2/2'$ doubly excited states can be classified in terms of the K , T and A quantum numbers which were used to describe the doubly excited states of a two-electron atom. For the $2/2'/2''$ triply excited states we have identified six resonances and their associated potential curves. Four of these six resonances are shown to be related approximately to the rotational excitations of a symmetric top formed by the three electrons. Finally, we point out that the present analysis is not complete. More accurate quantitative results would require the treatment of coupling among the adiabatic channels. Further understanding of the nature of correlations among the three electrons requires the analysis of the wavefunctions. Work along both directions is underway.

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