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Cold hydrogen–hydrogen scattering using CCA model

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Abstract. Cold hydrogen-hydrogen scattering has been investigated using close coupling approximation (CCA) model. The total wave function of the system is expanded in terms of atomic expansion basis. The effect of electron exchange and coupling to the continuum of both the atoms are taken into account. Singlet and triplet partial wave elastic and total cross-sections are presented and compared with existing theoretical predictions. Thermally averaged total cross-sections with respect to temperature are also provided along with their earlier results.

PACS. 34.50.-s Scattering of atoms and molecules – 31.15.Ar Ab initio calculations

1 Introduction

The development of low temperature physics initiates many investigations in different branches of physics. There was a revolution in experimental techniques for cooling and manipulating atoms using LASER based models that helps to achieve Bose Einstein Condensate (BEC) of atoms [1–4]. Investigations at cryogenic temperature have renewed interest in atom-atom scattering, in general. The low temperature scattering parameters for the H-H system have many applications in different branches of physics. It has been detailed by Koyama and Baird (KB) [5] and Jamieson et al. (JDY) [6]. In atom-atom collisions, the H-H scattering problem is the most simplest and the most detailed studied theoretically [5–13]. Moreover, the BEC of H atoms was successfully observed in 1998 by Fried et al. [4]. This has stimulated further studies on the H-H system. All the calculations have been carried out using the Born-Oppenheimer (B-O) separation model. In this model leptonic and hadronic motions are separated and the total wave function is expanded in a molecular orbital basis. This model is adiabatic in nature and the coupling between the nuclear and the light particles is neglected. One has to calculate the B-O potential at some discrete values for inter nuclear separation, R, and these values are then fitted to the long-range potentials $(-(\frac{C_6}{R^6}+\frac{C_8}{R^8}+\frac{C_{10}}{R^{10}}))$. It is very difficult to accurately calculate the B-O potential of an atom-atom system. The B-O potential, even for the H-H system, has been improved systematically over last three decades [14–20]. The effect of electron exchange in the asymptotic region is approximated by a local exchange potential. The theoreticians are not satisfied with their triplet B-O potential available so far for the H-H system. On the other hand, the singlet B-O potentials for hydrogen-alkali atom systems are not free from error.

Recently the B-O result for the H-H system is found to be extremely sensitive to the reduced mass of the system [22–24]. The effect on the results using nuclear and atomic mass is found to be differ by around 35%. Such dramatic change in the scattering length with mass factor has not been noticed earlier.

The present situation demands an alternative model to investigate atom-atom scattering. In our earlier attempt [33] we investigated H-H scattering using a close coupling model which is ab initio and non adiabatic in nature. We predicted the scattering lengths, effective ranges and thermally averaged total triplet cross-section at 2 K. As this was our first attempt we performed our calculations neglecting mass effect. In the close coupling approximation (CCA) the total wave function is expanded in terms of those of bound atomic subsystems, instead of using molecular orbital expansion employed earlier. The effect of exchange is explicitly taken care of by antisymmetrizing the total wave function of the system. The coupling between the constituent particles is included explicitly in this model. In this model one does not require to calculate the effective potentials (short and long ranges) separately. This model takes care of all the parts of the effective optical potentials automatically. The lowest order long range potential i.e. van der Waals potential and the higher order potentials are included via the basis sets employed in our model. We have included the effect of the continuum of both the atoms via pseudostates. This model is ab initio in nature and theoretically sound. The coefficients of the long range potential (C_6, C_8 and C_{10}) are found to be in good agreement with the exact values [33]. This is a completely different approach to the adiabatic model employed so far. Knowledge of the eigen and

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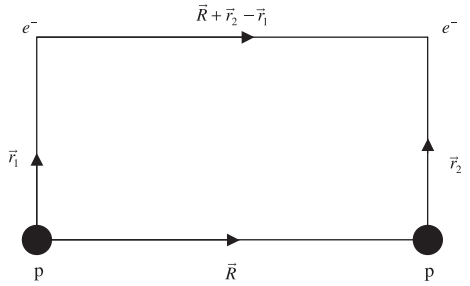

Fig. 1. The coordinate system employed.

pseudostates are sufficient to carry out the necessary calculations. This CCA model has been employed most successfully to anti-atom − atom scattering [26–28], positronium − atom [29–31] and positronium − positronium [32] scattering by our group.

In the present article we extend our earlier calculations having the same basis sets at higher energies and predict the results for few low order partial waves ($J = 0$ to 5) and total cross-sections for both the singlet and triplet states in the energy range 10^{-10} a.u. to 4×10^{-4} a.u. and compare with the existing theoretical predictions. We also compare our thermally averaged total triplet cross-section with the existing theoretical results as a function of temperature. This calculations are performed to find the suitability of the present model to investigate atom-atom scattering system.

2 Theory

In Close Coupling Approximation (CCA) method the total wave function of the system is expanded as

$$\psi^{\pm}(\vec{r_1},\vec{r_2},\vec{R}) = \frac{1}{\sqrt{2}}\sum(1\pm P_{12})\phi_\nu(\vec{r_1})\phi_\eta(\vec{r_2})F^{\pm}_{\nu\eta}(\vec{R}) \quad (1)$$

where P_{12} is the exchange operator. $\phi_\nu(\vec{r_1})$ and $\phi_\eta(\vec{r_2})$ stand for the wave functions describing the bound states of the colliding hydrogen atoms. In Figure 1, $\vec{r_1}$ and $\vec{r_2}$ are the position vectors of two electrons from their respective nucleus of each hydrogen atom and $\vec{R}$ is the hadronic separation. $F^{\pm}_{\nu\eta}(\vec{R})$ represents the unknown scattered wave function. The super script '+' stands for singlet scattering while '-' denotes triplet case. In the present model we assume that mass, M of the nucleus is infinitely heavy and the term of the order of $\frac{m}{M}$ has been neglected, m being the mass of the electron.

The total wave function ψ will satisfy the following Schrödinger equation

$$(H-E)\psi^{\pm}(\vec{r_1},\vec{r_2},\vec{R}) = 0. \quad (2)$$

One can recast the above equation into a set of coupled inhomogeneous integral equations for the scattering amplitudes in momentum space. Details of the deduction is given by Ghosh et al. [34]. The resulting three dimensional coupled integral equation (Lippmann-Schwinger type) for the scattering amplitude takes the form

$$f^{\pm}_{\nu'\eta';\nu\eta}(\vec{k}',\vec{k}) = f^{B\pm}_{\nu'\eta';\nu\eta}(\vec{k}',\vec{k}) - \frac{1}{2\pi^2}\sum_{\nu''\eta''}\int d\vec{k''}\frac{f^{B\pm}_{\nu'\eta';\nu''\eta''}(\vec{k}',\vec{k''})f^{\pm}_{\nu''\eta'';\nu\eta}(\vec{k''},\vec{k})}{k^2_{\nu''\eta''}-k''^2+i\epsilon} \quad (3)$$

where

$$f^{B\pm} = f^B \pm g^B \quad (4)$$

f^B and g^B are the first order direct and exchange scattering amplitudes respectively. Expression of the above scattering amplitudes, $f^{\pm}$ (Eqs. (3) and (4)) are exact. The direct (f^B) and exchange (g^B) scattering amplitudes are given by

$$f^B = -\frac{\mu}{2\pi}\int e^{iQR}\phi^*_{\eta'}(\vec{r_2})\phi^*_{\nu'}(\vec{r_1})V_d\phi_\nu(\vec{r_1})\phi_\eta(\vec{r_2})d\vec{r_1}d\vec{r_2}d\vec{R} \quad (5)$$

and

$$g^B = -\frac{\mu}{2\pi}\int e^{iQR}\phi^*_{\eta'}(\mid\vec{R}-\vec{r_1}\mid)\phi^*_{\nu'}(\mid\vec{R}+\vec{r_2}\mid) \times(H-E)\phi_\nu(\vec{r_1})\phi_\eta(\vec{r_2})d\vec{r_1}d\vec{r_2}d\vec{R}. \quad (6)$$

Here potential in the direct channel is

$$V_d = \frac{1}{R} - \frac{1}{\mid\vec{R}+\vec{r_2}\mid} - \frac{1}{\mid\vec{R}-\vec{r_1}\mid} + \frac{1}{\mid\vec{R}+\vec{r_2}-\vec{r_1}\mid}. \quad (7)$$

The evaluation of the direct first order matrix elements are straightforward. On the other hand, the exchange elements are very complicated as these involve multicentered integrals.

The total angular momentum, J, of the scattering system is a constant of motion. We express the scattering amplitude in partial wave decomposed form as

$$f^{\pm}_{\nu'\eta';\nu\eta}(\vec{k}',\vec{k}) = \frac{1}{\sqrt{kk'}}\sum_{JM}\sum_{J_1M_1}\sum_{J'_1M'_1}\sum_{LM_L}\sum_{L'M'_L}\begin{pmatrix}L' & l'_p & J'_1\\ M'_L & m'_p & M'_1\end{pmatrix} \times\begin{pmatrix}J'_1 & l'_t & J\\ M'_1 & m'_t & M\end{pmatrix}Y^*_{L'M'_L}(\hat{k}')T^{J\pm}(\tau'k';\tau k)Y_{LM_L}(\hat{k}) \times\begin{pmatrix}L & l_p & J_1\\ M_L & m_p & M_1\end{pmatrix}\begin{pmatrix}J_1 & l_t & J\\ M_1 & m_t & M\end{pmatrix} \quad (8)$$

and a similar expression for $f^{B\pm}$ with $T^{J\pm}$ on the right side of equation (8) is replaced by $B^{J\pm}$. Here l_p and l_t are the angular momenta of the bound projectile and the target atoms, respectively, and L represents the angular momentum of the moving hydrogen atom. τ represents the set of quantum numbers $(n_p, l_p, n_t, l_t, J_1, L)$. The corresponding final state quantum numbers are denoted by primes. After partial wave analysis, equation (3) becomes one-dimensional coupled-equation for unknown

amplitudes $T^{J\pm}$ as:

$$T^{J\pm}(\tau' k';\tau k) = B^{J\pm}(\tau' k';\tau k) - \frac{1}{2\pi^2}\sum_{\tau''}\int dk'' k'' \frac{B^{J\pm}(\tau' k';\tau'' k'')T^{J\pm}(\tau'' k'';\tau k)}{k^2_{\nu''\eta''} - k''^2 + i\epsilon}. \quad (9)$$

The real part of the elastic phase shift for Jth partial wave is given as:

$$\delta_J(k) = \frac{1}{2}\tan^{-1}\left\{\frac{\mathrm{Re}[T^J(\tau_0 k_0;\tau_0 k_0)]}{2\pi - \mathrm{Im}[T^J(\tau_0 k_0;\tau_0 k_0)]}\right\} \quad (10)$$

where τ_0 designates the initial states of both the atoms and the angle integrated Jth partial wave elastic cross section is given by

$$\sigma(J) = \frac{(2J+1)}{4\pi k_0^2}|T^J(\tau_0 k_0;\tau_0 k_0)|^2. \quad (11)$$

Here the total singlet and triplet cross-section is expressed as

$$\sigma_s(E) = \frac{1}{2}\sum_{even J}\sigma(J) + \frac{3}{2}\sum_{odd J}\sigma(J) \quad (12)$$

and

$$\sigma_t(E) = \frac{3}{2}\sum_{even J}\sigma(J) + \frac{1}{2}\sum_{odd J}\sigma(J) \quad (13)$$

respectively. The thermally averaged total cross-section (singlet or triplet) is defined by [6]

$$\langle\sigma_{(s/t)}(T)\rangle = \frac{1}{(k_B T)^2}\int_0^\infty \sigma_{(s/t)}(E) E \exp(-E/k_B T) dE \quad (14)$$

where k_B is Boltzmann's constant.

It is not possible to solve an infinite number of coupled equations (Eq. (9)). In practice, a few number of eigen or pseudo states are retained and the integral equations (Eq. (9)) are solved. The eigen and pseudostates are chosen such a way that should cover all the scattering space meaningfully.

In the present investigation, the following basis sets are used
(A) H($1s$)+H($1s$);
(B) H($1s, 2s, 2p$)+H($1s, 2s, 2p$);
(C) H($1s, 2s, 2\bar{p}$)+H($1s, 2s, 2\bar{p}$);
(D) H($1s, 2s, 2\bar{p}, 3\bar{d}$)+H($1s, 2s, 2\bar{p}, 3\bar{d}$).
The different basis sets have been employed to see the relative importance of each added eigen and pseudostates. Except from the first (A), which is known as static-exchange model, the other two models take care of the lowest order long range van der Waals interaction to different extent. The Model (D) includes higher order long range forces also. In model (C) and (D) we have used $2\bar{p}$ and $3\bar{d}$ pseudostates due to Damburg and Karule [35] which are of the form

$$\psi_{2\bar{p}}(\vec{r}) = \left(\frac{32}{129}\right)^{(\frac{1}{2})} r\left(1 + \frac{1}{2}r\right)\exp(-r) \quad (15)$$

and

$$\psi_{3\bar{d}}(\vec{r}) = \left(\frac{32}{535}\right)^{(\frac{1}{2})} r^2\left(\frac{1}{2} + \frac{1}{3}r\right)\exp(-r) \quad (16)$$

having binding energies

$$E_{2\bar{p}} = -\frac{7}{86}(\text{a.u.}) \quad (17)$$

and

$$E_{3\bar{d}} = \frac{13}{214}(\text{a.u.}). \quad (18)$$

3 Results and discussion

Two sets of coupled integral equations (Eq. (9)), one for singlet (T^{J^+}) and other for triplet (T^{J^-}), are solved numerically by matrix inversion method. Details of the method are given by Chakraborty et al. [36]. However, in the present case the convergence of the integral equation with respect to quadrature points is very complicated. We have to construct a new technique for the convergence of the integral equation. Sufficient care has been taken to achieve the convergent result. The error in the integration is less than 0.1%. Calculation has been performed for all the four basis sets mentioned above. This has been done to find the convergence of the results with added atomic states [33]. Scattering cross section decreases steadily from the static exchange value with the addition of higher excited states of both the atoms. We present the singlet scattering results with the use of basis set (C), as the predictions of the singlet obtained by using the basis set (D) are marginally different from the corresponding values using the basis set (C). On the other hand, it is essential to use the basis set (D) to get the scattering parameters for the triplet case. In atom-atom scattering the long range potentials are of key importance for the determination of scattering parameters. The lowest order long range potential is the van der Waals potential. In the present non adiabatic formalism the long range potentials are generated automatically. These values depend on the basis set employed. The first order values of the coefficients of the long range adiabatic potentials C_6, C_8 etc can also be estimated from our formalism [37]. The present estimated values of C_6, C_8 and C_{10} are given in our earlier paper [33] and found to be reliable. For the singlet case we use the basis set (C). It means that we have taken the lowest order van der Waals potential only. The effect of higher order potential has found to be marginal. This is also evident in our earlier paper [33] to study scattering lengths of this system. Scattering length is sensitive to the details of the potential. Table II of the earlier paper [33] shows that our singlet scattering length is converged with basis set (C) whereas higher order long range forces apart from van der Waals interaction is essentially required to converged result for triplet scattering length. Jamieson et al. [6] also mentioned this.

Here we present the results in the energy range 1×10^{-10} a.u. to 4×10^{-4} a.u. The s-wave singlet and triplet

Table 1. Partial wave cross-sections σ_J in units of 10^{-16} cm^2 for (a) singlet and (b) triplet as a function of relative energy E.

(a)	JDY [6]			Present		
E (10^{-6}) a.u.	$J=0$	$J=1$	$J=2$	$J=0$	$J=1$	$J=2$
0	0.59	0.00	0.00	1.12	0.00	0.00
1	2.83	0.30	0.00	3.48	0.057	0.0005
2	5.35	0.84	0.01	5.98	0.116	0.0038
3	7.93	1.37	0.02	8.43	0.138	0.010
4	10.4	1.80	0.05	10.79	0.125	0.021
5	12.8	2.10	0.08	13.0	0.091	0.033
6	15.1	2.26	0.12	15.1	0.050	0.050
7	17.2	2.31	0.17	17.0	0.016	0.066
8	19.1	2.25	0.23	18.8	0.0002	0.083

(b)	JDY [6]			Present		
E (10^{-6}) a.u.	$J=0$	$J=1$	$J=2$	$J=0$	$J=1$	$J=2$
0	12.6	0.00	0.00	16.61	0.00	0.00
1	17.3	0.21	0.00	19.43	0.079	0.0007
2	20.8	0.54	0.01	23.06	0.1892	0.0048
3	23.8	0.84	0.02	26.11	0.256	0.0136
4	26.5	1.05	0.05	28.73	0.274	0.028
5	28.7	1.16	0.08	31.01	0.252	0.048
6	30.8	1.19	0.12	33.0	0.203	0.073
7	32.5	1.15	0.17	34.76	0.143	0.103
8	34.1	1.05	0.23	36.29	0.082	0.136

Table 2. Thermally averaged total singlet and triplet cross-sections (10^{-16} cm^2) as functions of temperature.

	JDY [6]		Present	
Temperature T(K)	singlet	triplet	singlet	triplet
1	9.71	44.8	8.10	51.3
2	14.0	54.6	13.53	57.97
4	21.1	60.6	25.4	62.68
6	29.0	60.6	35.02	63.03
8	37.5	59.1	42.32	61.94

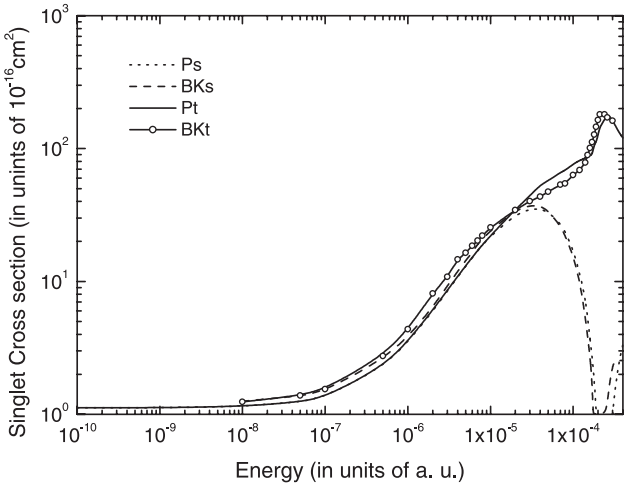

Fig. 2. Total and s-wave singlet scattering cross-sections (in units of 10^{-16}cm^2) for H−H scattering. Curves: Ps, Present s-wave cross-section; BKs, Koyama and Baird [5] s-wave cross-section; Pt, Present total cross-section; BKt, Koyama and Baird [5] total cross-section.

scattering lengths and effective ranges were reported and compared with existing theoretical predictions in our earlier paper [33].

Tables 1a and 1b represent our s-, p- and d-wave scattering cross-sections in the energy range 1×10^{-6} a.u. to 8×10^{-6} a.u. and are compared with those of Jamieson, Dalgarno and Yukich [6]. There are some differences between the present results and those of Jamieson et al. The present s-wave predictions for the singlet and triplet cases are higher than those of Jamieson et al. where as p- and d-wave cross-sections for both the cases are appreciably lower than those of the Harvard group. We hasten to add, the values of the d-wave cross-sections of Jamieson et al. for both the spin alignments are identical, where as we have obtained two sets of different results for singlet and triplet states. The differences between the present results and those of the Harvard group are not due to the asymptotic form of the long range potentials ($-\frac{C_6}{R^6}-\frac{C_8}{R^8}$) as values of C_6 and C_8 of present model differ marginally from those of the Harvard group. The present difference is due to the short range part of the potential employed. In B-O model, coupling between the nuclear and the light particles are neglected whereas present model includes it. The B-O potential is adiabatic in nature and in the present model short range potential generated is non adiabatic. Moreover, B-O potentials used by JDY are erroneous and ill conditioned which is evident from their later investigations.

In Table 2, we tabulate the present thermally averaged total cross-sections in the temperature range 1 to 8 K and compare with those of JDY [6]. Our values for singlet case are less than those of JDY up to the temperature 2 K and beyond this temperature the magnitude of the cross-sections of JDY are less than those of present model. In the case of triplet, the present thermally averaged cross-sections are always greater than those of JDY and the difference decreases with the increase of temperature. We have already mentioned the probable reason for these differences. As mentioned in our earlier paper [33], our triplet total thermally averaged cross-section at 2 K lies in between the maximum value of JDY and measured data [21].

Next we compare the present singlet total and partial wave cross-section in the energy range 1×10^{-10} to 4×10^{-4} a.u. with those of Koyama and Baird [5]. The total and s-wave cross-sections (Fig. 2) are in very good qualitative agreement with those of Koyama and Baird throughout the energy range considered. Peaks and positions of both the corresponding curves are nearly identical. However, the s-wave results of Koyama and Baird are greater than those of present predictions below 2×10^{-6} a.u. Figure 3 represents our p- and d-wave singlet cross-sections along with those of KB [5]. Here also we have noticed very good qualitative agreement. Positions and the values of the maxima of the present predictions for p- and d-wave cross-sections are almost equal to the corresponding predictions of KB. Similar feature has been obtained for $J=3,4,5$ partial cross-sections for singlet case (Fig. 4).

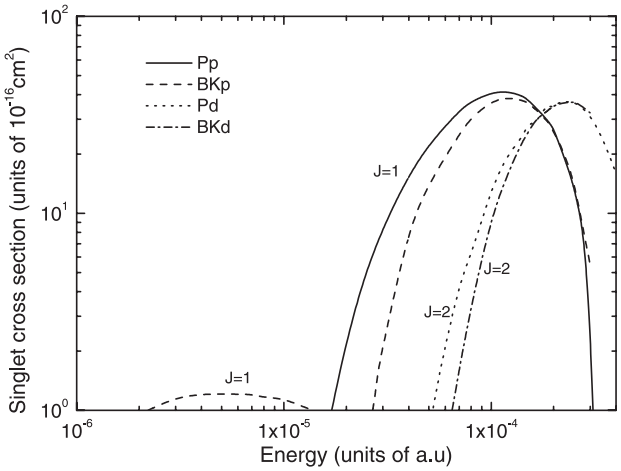

Fig. 3. The p-wave and d-wave singlet scattering cross-sections (in units of 10^{-16} cm^2) for H−H scattering. Curves: Pp, present p-wave cross-section; BKp, Koyama and Bairdé [5] p-wave cross-section; Pd, present d-wave cross-section; BKd, Koyama and Baird [5] d-wave cross-section.

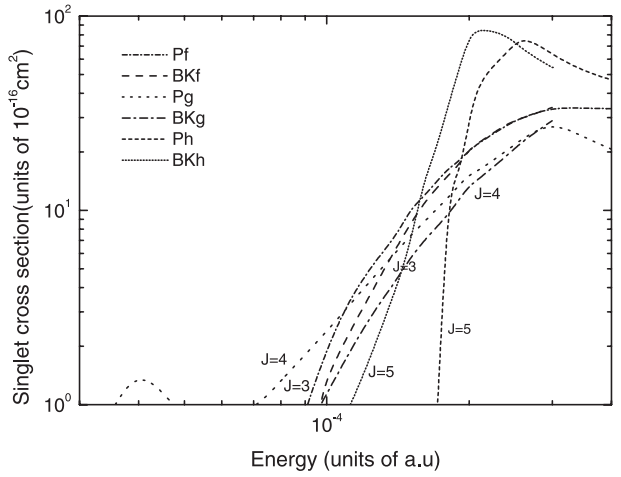

Fig. 4. The f-wave, g-wave and h-wave singlet scattering cross-sections (in units of 10^{-16} cm^2) for H−H scattering. Curves: Pf, present f-wave cross-section; BKf, Koyama and Baird [5] f-wave cross-section; Pg, present g-wave cross section; BKg, Koyama and Baird [5] g-wave cross-section; Ph, present h-wave cross-section; BKh, Koyama and Baird [5] h-wave cross-section.

We present total and partial wave cross-sections for triplet state in Figure 5. The total elastic triplet cross-section is obtained by adding the first six partial wave cross-sections. We have calculated higher partial waves and there is no contribution of them in the above mentioned energy range. Since previous investigations for individual triplet partial waves cross-sections in this energy range (1×10^{-10} to 4×10^{-4} a.u.) have not been reported, we cannot compare our results with any other existing results. The present total triplet cross-sections are appreciably greater than that of Koyama and Baird below the incident energy 1×10^{-5} a.u. and beyond that our total

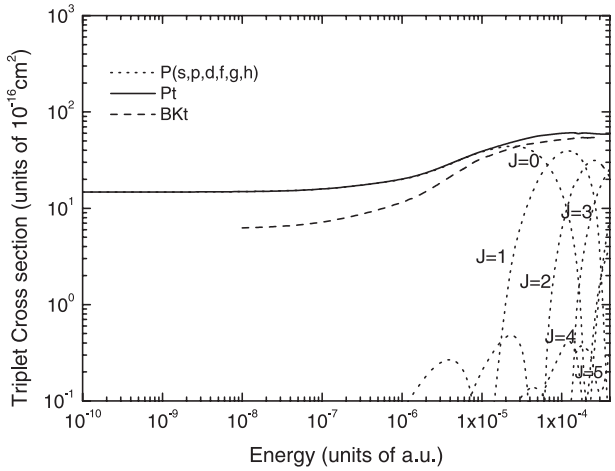

Fig. 5. Triplet scattering cross-sections (in units of 10^{-16} cm^2) for H−H scattering. Curves: Pt, present total cross-section; BKt, Koyama and Baird [5] total cross-section; P(s, p, d, f, g, h) curves are for present partial wave cross-sections from s- to h-waves.

cross-section are more or less similar to those of Koyama and Baird.

4 Conclusion

In the present work we have employed close coupling models to investigate H-H scattering in the energy range 1×10^{-10} to 4×10^{-4} a.u. Our model is ab initio and non adiabatic in nature. The accuracy of the prediction depends on the basis set employed. Four different basis sets have been used to investigate the convergence of the results with added eigen and pseudostates. The present basis set (model D) represents the total effective singlet and triplet potentials in a satisfactory way [33] and this potential generated by the present model is unique for the particular basis set.

It has been pointed out by the Harvard group and others that non adiabatic effect is important in studying cold atom-atom scattering. Jamieson and Dalgarno [23] found that non adiabatic correction to the adiabatic one for the singlet scattering length is by about 25%. On the other hand our model is purely non adiabatic in nature.

The present model is completely different from the adiabatic one as one does not need to calculate the potentials separately. The present scattering parameters, obtained here, are in satisfactory agreement with the standard adiabatic results of JDY [6] and KB [5]. There is marginal differences for each partial waves (from $J = 0$ to 5) singlet cross-sections between ours and those of KB. These differences are due to the short range part of their B-O potentials which are not accurate and due to neglect of non adiabatic effect.

In the present calculation we have not taken into account the reduced mass effect. However, in a static exchange model we perform the calculation including the mass effect. The results differ by less than 2%. We have

included the effect of the continuum of both the atoms via pseudostates. Here we demonstrate the suitability of the CCA model to investigate H-H scattering. The accuracy of the present model depends on the scattering space included in the calculation. The present results may be refined by using even more elaborate basis sets slightly. The effect of reduced mass may change the present results to some extent. The application of the present model to other atomic systems is rather straight-forward. We advocate this non adiabatic model to study atom-atom scattering system at low energies.

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