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Doubly excited $^3S^e$, $^3D^e$ and $^3G^e$ states of two-electron atomic systems

A. K. Das, T. K. Ghosh, P. K. Mukherjee

Department of Spectroscopy, Indian Association for the Cultivation of Science, Jadavpur,
Calcutta 700 032, India

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Summary. Time-dependent perturbation theory has been applied to calculate the doubly excited triplet states *Nsns*: $^3S^e$, *Npnp*: $^3D^e$ and *Ndnd*: $^3G^e$ ($N = 2, 3, 4$, $n = N + 1, \dots, 5$) for He, Li^+, Be^{2+} and B^{3+}. A time-dependent harmonic perturbation causes simultaneous excitation of both the electrons with a change of spin state. The doubly excited energy levels have been identified as the poles of an appropriately constructed linearized variational functional with respect to the driving frequency. In addition to the transition energies, effective quantum numbers of these doubly excited states have been calculated and analytic representations of their wave functions are obtained. These are utilized to estimate the Coulomb repulsion term for these states which checks the consistency of the wave functions. These wave functions may also be used for calculating other physical properties of the systems.

Key words: Doubly excited states – Excitation energies – Effective quantum numbers

1 Introduction

Study of the doubly excited states of atoms has growing interest during the last two decades chiefly because of their importance in interpreting astrophysical data [1, 2] and their level description in terms of a new set of quantum numbers [3–7] which represents a complete departure from conventional single particle picture [8]. Although it is observed first in helium by Madden and Codling [9] in photoabsorption experiments, now it is experimentally observed for highly charged ions also [10].

Theoretical calculations so far done involve two distinct approaches. The first one involves collisional methods as pioneered by Burke and Taylor [11], O'Malley and Geltman [12], Macek [13] and more recently by Callaway [14], Bhatia and Temkin [15], Lipsky et al. [16], Chung and Davis [17]. In their analysis the doubly excited states are treated as quasi-bound resonances embedded in scattering continuum. The other approach is based on treating the levels from bound state point of view. To this end accurate calculations using complex rotation method by Ho and co-workers [18], hyperspherical co-ordinate approach by Lin [19] and others [20–24] and several other approaches [25, 26] are important, especially the molecular orbital (MO) approach by Faegin and Briggs [27] to find these resonant states. The method was reviewed by Rost and Briggs [28] recently which gives a clear picture of the origin of the quantum numbers described by Herrick et al. [4]. The distinguishing characteristics like excitation and decay of doubly excited states of atoms and ions have been discussed thoroughly by Rau [29].

Experimental methods use photo-absorption techniques [9], electron and ion impact observation [30] and the more recent beam-foil techniques [31] and ejected electron spectra [32, 33]. Good reviews are now available on such calculations [34].

In the present communication time-dependent perturbation theory has been applied to calculate the doubly excited states *Nsns*: $^3S^e$, *Npnp*: $^3D^e$ and *Ndnd*: $^3G^e$ ($N = 2, 3, 4$, $n = N + 1, \ldots, 5$) for the helium isoelectronic ions up to B^{3+}. The method which we adopted is based on bound state approach. The method was discussed earlier in detail by Mukherjee and co-workers [35–37] and applied for studying the doubly excited triplet transitions *Nsnp*: $^3P^o$, *Nsnd*: $^3D^e$ and *Npnd*: $^3F^o$ from $^1S^e$ ground state of helium isoelectronic sequence [38]. We seek the natural excitation modes of the two electron charge cloud using a correlated description of the electron pair. A suitable form of two-particle harmonic perturbation is chosen which induces simultaneous excitations of the electrons to different spatially excited states accompanied by a change in the spin state. In Sect. 2 we give a brief description of the method. Section 3 will deal with a discussion of the present results.

2 Method

Let us consider the time evolution of a two electron system, initially at the ground state Ψ with energy E_0 of the usual non-relativistic Hamiltonian H_0, subject to a spin-dependent harmonic perturbation of the form

$$H'(\boldsymbol{r}, \boldsymbol{\sigma}, t) = G(\boldsymbol{r}, \boldsymbol{\sigma})e^{-i\omega t} + G^+(\boldsymbol{r}, \boldsymbol{\sigma})e^{i\omega t}. \tag{1}$$

The perturbation $G(\boldsymbol{r}, \boldsymbol{\sigma})$ is such that it excites both the electrons simultaneously to a new state changing the spin multiplicity from singlet to triplet. A suitable choice of the perturbation may be

$$G(\boldsymbol{r}, \boldsymbol{\sigma}) = \lambda[h(\boldsymbol{r}_1)h'(\boldsymbol{r}_2) - h(\boldsymbol{r}_2)h'(\boldsymbol{r}_1)]\ [S_-(1)S_+(2) - S_-(2)S_+(1)], \tag{2}$$

where λ is the perturbation strength parameter, S_+ and S_- are the usual spin up and spin down operators. The spatial term excites the electrons from the ground state. The general structure of the one particle term is given by

$$h(\boldsymbol{r}) = r^l P_l(\cos\theta), \tag{3}$$

$l = 0, 1, 2, \ldots$, will excite the ground s orbital to $s, p, d, \ldots$, symmetries respectively.

Following [38] we can write the time evolution of the total wave function as

$$\Phi(t) = N[\Psi + \delta\Psi^- e^{-i\omega t} + \delta\Psi^+ e^{i\omega t}]\, e^{-iEt}, \tag{4}$$

where $\delta\Psi^\pm$ are the first order admixtures to Ψ due to two harmonic components of the perturbation. Here the spin part of Ψ and $\delta\Psi$ are different. N is a normalization constant to be determined from the condition [38]

$$\frac{1}{T}\int_0^T \langle\Phi|\Phi\rangle\, dt = 1. \tag{5}$$

Since the perturbing Hamiltonian equation (2) changes the spin multiplicity, the spin part of the Ψ and $\delta\Psi^{\pm}$ are different. To determine the time evolution of the total wave function we have to determine the first order perturbed functions $\delta\Psi^{\pm}$. These are obtained by constructing a time averaged functional

$$J(\Phi) = \frac{1}{T}\int_0^T \langle\Phi|H_0 + H' - \mathrm{i}\frac{\partial}{\partial t}|\Phi\rangle\,\mathrm{d}t \tag{6}$$

subject to the optimization condition

$$\delta J(\Phi) = 0, \tag{7}$$

with respect to parameters introduced in the functions $\delta\Psi^{\pm}$. We expand the spatial part of $\delta\Psi^{\pm}$ in the following manner:

$$\delta\Psi^{\pm} = \sum C_i^{\pm}\,\eta_i(1,2), \tag{8}$$

where $\eta_i(1,2)$ are correlated pair bases formed out of one particle Slater type orbitals (STOs) [38]

$$\eta_i(1,2) \sim \xi_k(1)\,\xi_l'(2) - \xi_k(2)\,\xi_l'(1) \tag{9}$$

and $C_i^{\pm}$ are the linear variation parameters. Choice of exponents of the Slater bases depends on the symmetry and principal quantum number of the excited orbitals and are preassigned here. We expand the functional [Eq. (6)] in terms of $\delta\Psi^{\pm}$ and retain terms up to quadratic in $\delta\Psi^{\pm}$. The optimization condition, namely

$$\frac{\partial J(\Phi)}{\partial C^{\pm}} = 0, \tag{10}$$

leads to sets of decoupled linear equations in the unknown parameters $C^{\pm}$ [38] which can readily be solved for a given external frequency to get the response characteristics of the system. A discussion of the results is given in the next section.

3 Results and discussions

Low and moderately high lying doubly excited triplet transitions viz $1s^2$: $^1\mathrm{S}^e \rightarrow Nsns$: $^3\mathrm{S}^e$; $Npnp$: $^3\mathrm{D}^e$ and $Ndnd$: $^3\mathrm{G}^e$, $N = 2, 3, 4$ and $n = N + 1, \ldots, 5$ have been studied for helium isoelectronic sequence up to B^{3+}. For He we used radially correlated ground state wave function of Mukherji [39] computed with Weiss [40] exponents. For Li^{+}, Be^{2+} and B^{3+} the ground state wave functions are obtained from Mukherji [39] computed with Clementi [41] exponents. For all the perturbed orbitals 7 parameter STO representation has been used. The choice of the set of exponents of the perturbed orbitals depends on the principal quantum number and the symmetry of the orbital. In the present triplet state calculation although the angular momentum of the orbitals are the same, the inner and outer principal quantum numbers are different. Accordingly we have chosen different basis sets for the inner and outer orbital representations. We do not mix basis sets of different symmetries and thus take care of radial correlation only. The parameters of the product basis set are determined, through optimization procedure at each external frequency and provide very reasonable description of the perturbed functions at the given frequency. The optimized functional $J(\Phi)$ rises monotonically with respect to the driving frequency ω with occasional real poles. The pole positions determine doubly excited modes of the unperturbed Hamiltonian and hence positions of these poles represent the doubly excited energy levels measured from the ground state energy of the systems. The first order perturbed function $\delta\Psi^-$ blows up at pole positions and renormalization of the first order perturbed function at the poles furnishes adequate description of the doubly excited wave functions [38].

In Table 1 the calculated TDVPT $^3S^e$ double excitation energies are displayed along with the values of the Coulomb repulsion integral in the excited state for He, Li^+, Be^{2+} and B^{3+}. The level description was done according to the configuration scheme of Cooper et al. [8]. Such results for the triplet excitations to $^3D^e$ and $^3G^e$ states for all the ions have been listed in Tables 2 and 3, respectively. In all cases transition energy was measured from the ground state $^1S^e$. We have chosen the angular part of the two particle perturbation operator in such a way that *Npnp*: $^3D^e$ and *Ndnd*: $^3G^e$ final states are allowed. The quantum numbers N, K, T, n and A of the doubly excited states as prescribed by the group theoretical analysis of Wulfman [3], Herrick and Sinanoglu [4] and Lin [7, 19] are also listed in the respective tables to have correspondence with the configuration state description. In the group theoretical analysis effect of radial and angular correlations have been taken care of, whereas our method incorporates radial correlations only. The effect of radial and angular correlations on the doubly excited states was thoroughly discussed by Lin [7] and Ezra and Berry [42] using hyperspherical co-ordinate method.

Most of the theoretical data available on transition energy were compiled by Shearer-Izumi [43]. In this regard Lipski et al. [16] have done important and exhaustive calculations using configuration interaction (CI). We compared our data with those compiled by Shearer-Izumi [43], Lipski et al. [16] and with the accurate results using complex rotation method by Ho and co-workers [18]. Experimental data are available only for very few of the triplet transitions $^3D^e$ [44, 45] and we have accordingly listed them in Table 2. Theoretical data existing for the $^3G^e$ states are rather scanty. Available data are listed in Table 3. Most of the theoretical calculations use highly correlated basis sets for both the ground as well as excited states, hence the data are very accurate. As discussed in [38], the discrepancy of our results arises from two sources. Firstly the reference point, namely the ground state energy from where the excitation energy is measured is taken from the radially correlated calculation of Mukherji [39] (results with superscript α in the tables). These give a little underestimate of the transition energies. In order to rectify this error we have also chosen the reference point as that given by Pekeris [46]. Results with superscript β in the tables are with these reference ground state energies. The second source of errors comes from the neglect of angular correlation in the excited states. From a close look at the calculated and existing accurate data listed in Tables 1–3 we find that the maximum deviation which occurs for He is about 0.7% and the deviation diminishes as we go towards transitions of higher inner or outer quantum numbers or transitions of higher isoelectronic members. This is quite reasonable since the effect of interelectronic correlation is relatively less for such transitions. The overall agreement of the computed data with the existing ones clearly indicates that the effect of angular correlations in the excited states is confined to within 1% for all the systems under study. Along with the transition energies we get analytic wave functions for the doubly excited states in terms of Slater bases. These analytic wave functions contain lesser number of parameters than those from traditional variational calculations. These wave functions may be useful for calculating other properties like the autoionisation rates, transition probabilities etc. In the present case we have calculated the expectation value of the Coulomb repulsion term in the doubly

Table 1. Energies (measured from ground state), Coulomb repulsions and effective quantum numbers (n^*) of the doubly excited $^3S^e$ states below the $N = 4$ hydrogenic threshold of two-electron atomic systems

Ion	(Nl, nl)	$N(K,T)_n^A$	Excitation energy (a.u.)			n^*		Coulomb repulsion (a.u.)
			Present[α]	Present[β]	Other values	Present	Others	
He	2s3s	$2(1,0)_3^-$	2.2941	2.3188	2.3018[a], 2.3017[b], 2.3012[c]	2.4254	2.2129[b]	0.1187
	2s4s	$2(1,0)_4^-$	2.3370	2.3617	2.3508[a], 2.3553[b], 2.3442[c]	3.4462	3.2094[b]	0.0672
	2s5s	$2(1,0)_5^-$	2.3538	2.3785	2.3762[a], 2.3756[b], 2.3550[c]	4.4455	4.2076[b]	0.0423
	3s4s	$3(2,0)_4^-$	2.6067	2.6314	2.6172[a], 2.6166[b], 2.6165[c,d]	3.1560	2.7740[b]	0.0667
	3s5s	$3(2,0)_5^-$	2.6284	2.6531	2.6465[a], 2.6458[b], 2.6457[c,d]	4.1885	3.7362[b]	0.0428
	4s5s	$4(3,0)_5^-$	2.7226	2.7473		3.9841		0.0460
Li^+	2s3s	$2(1,0)_3^-$	5.8417	5.8695	5.8411[b]	2.6449	2.5224[b]	0.1985
	2s4s	$2(1,0)_4^-$	5.9782	6.0060	5.9982[b]	3.6588	3.5271[b]	0.1172
	2s5s	$2(1,0)_5^-$	6.0354	6.0632	6.0580[b]	4.6575	4.5318[b]	0.0786
	3s4s	$3(2,0)_4^-$	6.5828	6.6106	6.5912[b]	3.4320	3.2511[b]	0.1551
	3s5s	$3(2,0)_5^-$	6.6541	6.6819	6.6690[b]	4.5061	4.2374[b]	0.0745
	4s5s	$4(3,0)_5^-$	6.8640	6.8918		4.3163		0.0691
Be^{2+}	2s3s	$2(1,0)_3^-$	11.0289	11.0583	11.0193[b]	2.7411	2.6560[b]	0.2760
	2s4s	$2(1,0)_4^-$	11.3077	11.3371	11.3215[b]	3.7494	3.6611[b]	0.1667
	2s5s	$2(1,0)_5^-$	11.4279	11.4573	11.4506[b]	4.7446	4.6668[b]	0.1104
	3s4s	$3(2,0)_4^-$	12.3950	12.4244	12.3922[b]	3.6173	3.4595[b]	0.1505
	3s5s	$3(2,0)_5^-$	12.5302	12.5596	12.5413[b]	4.6435	4.4521[b]	0.1038
	4s5s	$4(3,0)_5^-$	12.9061	12.9355		4.5053		0.1023
B^{3+}	2s3s	$2(1,0)_3^-$	17.5853	17.8855	17.8379[b]	2.7936	2.7309[b]	0.3531
	2s4s	$2(1,0)_4^-$	18.3251	18.3553	18.3373[b]	3.7956	3.7354[b]	0.2161
	2s5s	$2(1,0)_5^-$	18.5308	18.5610	18.5546[b]	4.7836	4.7405[b]	0.1436
	3s4s	$3(2,0)_4^-$	20.0469	20.0771	20.0214[b]	3.7477	3.5769[b]	0.3456
	3s5s	$3(2,0)_5^-$	20.2539	20.2841	20.2640[b]	4.6971	4.5720[b]	0.1333
	4s5s	$4(3,0)_5^-$	20.8461	20.8763		4.6001		0.1248

Excitation energies using correlated ground state of [α] Mukherji [39] and [β] Pekeris [46], [a] Ref [43], [b] Ref [16], [c] Ref [18], [d] Ref [14]

Table 2. Energies (measured from ground state), Coulomb repulsions and effective quantum numbers (n^*) of the doubly excited $^3D^e$ states below the $N = 4$ hydrogenic threshold of two-electron atomic systems

Ion	(Nl, nl)	$N(K,T)_n^A$	Excitation energy (a.u.)			n^*		Coulomb repulsion (a.u.)
			Present$^\alpha$	Present$^\beta$	Other values	Present	Others	
He	2p3p	$2(1,0)_3^-$	2.3049	2.3296	2.3217^a, 2.3206^b, 2.3196^e, 2.3206^f	2.5959	2.4515^b	0.1204
	2p4p	$2(1,0)_4^-$	2.3408	2.3655	2.3627^a, 2.3624^b, 2.3613^e, 2.3623^f	3.6131	3.4764^b	0.0661
	2p5p	$2(1,0)_5^-$	2.3556	2.3803	2.3800^a, 2.3788^b	4.6127	4.4852^b	0.0419
	3p4p	$3(0,0)_4^-$	2.6113	2.6360	2.6441^a, 2.6423^b, 2.6367^d	3.3113	3.5680^b	0.0680
	3p5p	$3(0,0)_5^-$	2.6332	2.6579	2.6596^a, 2.6583^b, 2.6584^d	4.5932	4.6316^b	0.0530
	4p5p	$4(-1,0)_5^-$	2.7244	2.7491	2.7544^a, 2.7526^e	4.1030		0.0470
Li$^+$	2p3p	$2(1,0)_3^-$	5.8625	5.8903	5.8764^b	2.7467	2.6774^b	0.2102
	2p4p	$2(1,0)_4^-$	5.9863	6.0141	6.0094^b	3.7622	3.7017^b	0.1206
	2p5p	$2(1,0)_5^-$	6.0394	6.0672	6.0654^b	4.7619	4.7141^b	0.0780
	3p4p	$3(0,0)_4^-$	6.5957	6.6235	6.6376^b	3.5703	3.7425^b	0.1145
	3p5p	$3(0,0)_5^-$	6.6580	6.6858	6.6934^b	4.5980	4.7934^b	0.0755
	4p5p	$4(-1,0)_5^-$	6.8684	6.8962		4.4070		0.0720
Be^{2+}	2p3p	$2(1,0)_3^-$	11.0594	11.0888	11.0712^b	2.8137	2.7711^b	0.2979
	2p4p	$2(1,0)_4^-$	11.3201	11.3495	11.3442^b	3.8242	3.7918^b	0.1743
	2p5p	$2(1,0)_5^-$	11.4341	11.4635	11.4622^b	4.8200	4.8051^b	0.1136
	3p4p	$3(0,0)_4^-$	12.4072	12.4366	12.4597^b	3.8187	3.6835^b	0.1600
	3p5p	$3(0,0)_5^-$	12.5387	12.5681	12.5778^b	4.7410	4.8600^b	0.1158
	4p5p	$4(-1,0)_5^-$	12.9123	12.9417		4.5696		0.1035
B^{3+}	2p3p	$2(1,0)_3^-$	17.8956	17.9258	17.9064^b	2.8502	2.8225^b	0.3849
	2p4p	$2(1,0)_4^-$	18.3418	18.3720	18.3679^b	3.8540	3.8394^b	0.2278
	2p5p	$2(1,0)_5^-$	18.5392	18.5694	18.5706^b	4.8422	4.8516^b	0.1493
	3p4p	$3(0,0)_4^-$	20.0512	20.0814	20.1098^b	3.7619	3.8602^b	0.2892
	3p5p	$3(0,0)_5^-$	20.2621	20.2923	20.3127^b	4.7511	4.8938^b	0.1413
	4p5p	$4(-1,0)_5^-$	20.8526	20.8828		4.6400		0.1280

Excitation energies using correlated ground state of $^\alpha$ Mukherji [39] and $^\beta$ Pekeris [46], ^{a} Ref [43], ^{b} Ref [16], ^{c} Ref [18], ^{d} Ref [14], ^{e} Expt. value Ref [45], ^{f} Expt. value Ref [44]

Table 3. Energies (measured from ground state), Coulomb repulsions and effective quantum numbers (n^*) of the doubly excited $^3G^e$ states below the $N = 4$ hydrogenic threshold of two-electron atomic systems

Ion	(Nl, nl)	$N(K,T)_n^A$	Excitation energy (a.u.)			n^*		Coulomb repulsion (a.u.)
			Present[α]	Present[β]	Other values	Present	Others	
He	$3d4d$	$3(1,1)_4^0$	2.6160	2.6407	2.6447[c,d]	3.4964	3.6811[c,d]	0.0681
	$3d5d$	$3(1,1)_5^0$	2.3655	2.6602	2.6623[d]	4.8337	5.0900[d]	0.0541
	$4d5d$	$4(0,1)_5^0$	2.7258	2.7505	2.7528[c]	4.2033	4.3853[c]	0.0468
Li^+	$3d4d$	$3(1,1)_4^0$	6.6049	6.6327		3.6798		0.1195
	$3d5d$	$3(1,1)_5^0$	6.6623	6.6901		4.7062		0.0773
	$4d5d$	$4(0,1)_5^0$	6.8714	6.8902		4.2845		0.0750
Be^{2+}	$3d4d$	$3(1,1)_4^0$	12.4208	12.4502		3.7612		0.1696
	$3d5d$	$3(1,1)_5^0$	12.5418	12.5712		4.7782		0.1121
	$4d5d$	$4(0,1)_5^0$	12.9203	12.9497		4.6569		0.1096
B^{3+}	$3d4d$	$3(1,1)_4^0$	20.0635	20.0937		3.8035		0.2192
	$3d5d$	$3(1,1)_5^0$	20.2707	20.3009		4.8100		0.1464
	$4d5d$	$4(0,1)_5^0$	20.8654	20.8956		4.7223		0.1626

Excitation energies using correlated ground state of [α] Mukherji [39] and [β] Pekeris [46], [c] Ref [18], [d] Ref [14] excited state. The very regular behavior of the expectation values as reflected from the tables, show the consistency of the excited state wave functions. These wave functions may be effectively used for collision calculations involving doubly excited states, many of whom, are important in the solar chromosphere.

As an extra check for the doubly excited energy levels we estimated the effective quantum numbers n^* of such states using the formula [16]

$$\varepsilon = -\frac{1}{2}\left[\left(\frac{Z}{N}\right)^2 + \left(\frac{Z-1}{n^*}\right)^2\right] \tag{11}$$

where ε is the energy of the doubly excited state (in a.u.) measured from the ionisation threshold, N is the principal quantum number of the inner electron and Z is the nuclear charge. The calculated n^* values have been compared with those obtained by Lipsky et al. [16] in Tables 1, 2 for $^3S^e$ and $^3D^e$ states, while in Table 3 for the few $^3G^e$ states these are compared using the existing data of Callaway [14] and Ho [18]. The percentage deviation is maximum for the lowest transition in He and it diminishes as we move towards higher lying excitations. This feature is reflected for all the ions. The deviation also diminishes as we move along higher members of the isoelectronic sequence. The effective quantum number n^* depends sensitively on the energy levels. The relatively large difference of our n^* values from those obtained from experimental data is not due to the present methodology but is coming from the choice of our radially correlated basis sets. Results are expected to be more accurate using basis sets which include radial as well as angular correlations. In the present scheme we calculated the transitions only up to $N = 4$ and $n = 5$ levels. As for higher lying transitions, particularly for $^3G^e$ states practically no data is available for comparison. Angular correlation may be incorporated in these calculations by suitably extending the product basis sets. This is under present consideration and results will be reported in due course.

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