

Energy, fine structure, and hyperfine structure of the core-excited states $1s2s2pnp$ 5P ($n = 2-5$) and $1s2p^2mp$ 5S ($m = 2-5$) for Li^- ion

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Abstract. The relativistic energies, the oscillator strength, and the lifetimes of high-lying core-excited states $1s2s2pnp$ 5P ($n = 2-5$) and $1s2p^2mp$ $^5S^o$ ($m = 2-5$) of Li^- ion are calculated with the saddle-point variational method and restricted variation method. The fine structure and the hyperfine structure of the core-excited states for this system are also explored. The results are compared with other theoretical and experimental data in the literature.

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1 Introduction

Recently, the core-excited states of beryllium-like ions, a complex four-electron atomic system, have been of interest to both theorists [1,8–15] and experimentalists [2–7]. The studies of the core-excited states of beryllium-like ions are very useful in astrophysics and plasma physics. The structures of the core-excited states of Li^- ions provide important information on the dynamics of many-electron interactions, as the weakly bounded electrons are easily perturbed.

Bunge [1,2] first revealed the existence of two core-excited bound states of Li^- and identified the 3490 Å line as the $1s2s2p^2$ $^5P - 1s2p^3$ 5S transition in Li^- . The results of Bunge were in good agreement with the experiment of Berry et al. [3] and were readily confirmed experimentally [5–7]. Brooks and his group [6] identified the transition $1s2s2p^2$ $^5P - 1s2p^3$ $^5S^o$ for Be-like isoelectronic series from Li^- to F_{VI} . In 1983, Beck and Nicolaides [8] reported the lifetime and the hyperfine structure of the doubly excited states of $1s2s2p^2$ 5P and $1s2p^3$ 5S for Li^- ion in a many-body point of view, they proposed the additional decay channel of radiative autoionization similar to Li [9] and obtained a theoretical lifetime 2.7 ns of $^5S^o$ state. The fine structure and the hyperfine structure are sensitive to the correlation effects between electrons and the relativistic corrections. The hyperfine structure is caused by the interaction between the electrons and the electromagnetic multipole moments of the nucleus. The Li^- ion is known to support a sharp E1 transition line between the $1s2s2p^2$ $^5P_{1,2,3}$ and the $1s2p^3$ $^5S^o$ states

at 3489.7 Å from Bunge [1]. Cheng et al. [10] reported the fine and the hyperfine structure of the doubly excited quintet states of the $^7\text{Li}^-$ spectrum using multiconfiguration Dirac-Fock method (MCDF). More recently, Yang and Chung [11] reported the results of energy, fine structure, and hyperfine structure of the lowest core-excited states of $1s2s2p^2$ 5P and $1s2p^3$ $^5S^o$ of the Be-like isoelectronic sequence ($Z = 3-10$) using multiconfiguration interaction method. So far, to our knowledge, no calculations have been reported for the high- n core-excited states of the Li^- ion. The theoretical calculations on the structure of high- n core-excited states for the four-electron negative ions are complicated. The negative ion is very unstable and the weakly bound electrons are easily perturbed. The inner three electrons exert an efficient screening upon the nucleus. With the increase of the parameter n , the screening grows more and more obvious, and the calculations are challengeable. On the other hand, the identification of the transition line among high- n core-excited quintet states is very difficult, due to the restriction of resolution from experiments.

In this paper, the saddle-point variational method is used on the high-lying core-excited states $1s2s2pnp$ 5P ($n = 2-5$) and $1s2p^2mp$ $^5S^o$ ($m = 2-5$) for Li^- . The non-relativistic energies, the fine structure, the hyperfine parameters and the coupling constants are reported. Relativistic effects are computed with first-order perturbation theory. The oscillator strengths, lifetimes and wavelength are also reported to compare with experimental and theoretical data. These available data should be very useful in the better understanding of the experimental spectra in the future.

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2 Theory

In LS coupling, the non-relativistic Hamiltonian for the Be-like system is given by

$$\hat{H}_0 = \sum_{i=1}^4 \left[-\frac{1}{2} \nabla_i^2 - \frac{4}{r_i} \right] + \sum_{\substack{i,j=1 \\ i < j}}^4 \frac{1}{r_{ij}}. \quad (1)$$

The saddle-point wave function ψ_b of the core-excited four-electron system can be written as

$$\psi_b(1, 2, 3, 4) = A \sum_i C_i [1 - P(r_i)] \phi_{n(i), l(i)}(R) Y_{l(i)}^{LM}(\Omega) \chi_{SM_S} \quad (2)$$

where A is the antisymmetrization operator and the radial basis function is Slater orbital

$$\phi_{n(i), l(i)}(R) = \prod_{j=1}^4 r_j^{n_j} \exp(-\alpha_j r_j). \quad (3)$$

In equation (2), the projection operator is given by

$$P(r_i) = |\phi_0(r_i)\rangle \langle \phi_0(r_i)|. \quad (4)$$

Here the vacancy orbital is

$$\phi_0(r) = N \exp(-qr) \quad (5)$$

where q is a parameter determined in the energy maximization processes; the linear parameters C_i and the nonlinear parameters α_j in equation (3) are determined in the energy optimization processes.

A different set of α_j is used for each $l(i)$. The angular and spin wave function can be represented as

$$l(i) = [(l_1, l_2) l_{12}, l_3] l_{123}, l_4 \quad (6)$$

$$\chi_{SS_z} = [(s_1, s_2) s_{12}, s_3] s_{123}, s_4. \quad (7)$$

We use restricted variational method to further improve energy E_b . The basic wave function ψ_b is used as a single term in the improved wave function [12], that is expressed as

$$\Phi(1, 2, 3, 4) = D_0 \psi_b(1, 2, 3, 4) + \psi_2(1, 2, 3, 4) \quad (8)$$

where

$$\psi_2(1, 2, 3, 4) = A \sum_{i=1}^I D_i \Phi_{n(i)l(i)}(1, 2, 3, 4). \quad (9)$$

To saturate the functional space, ψ_2 takes the same form as $\psi_b(1, 2, 3, 4)$, but the nonlinear parameters are much different from those of $\psi_b(1, 2, 3, 4)$. Each of the nonlinear parameters in the basis function of ψ_2 is optimized in the restricted variational calculation and the energy improvement ΔE_{RV} can be obtained. Then the nonrelativistic energy is given by $E_b + \Delta E_{RV}$.

In addition to the ΔE_{RV} , the energy is further corrected by the mass polarization effect and relativistic corrections. The relativistic perturbation operators considered in this work are: correction to the kinetic energy (P^4), the Darwin term, the electron-electron contact term, and the orbit-orbit interaction. The mass polarization perturbation operator is accurate to all orders. The relativistic corrections are calculated using the first-order perturbation theory [13]. The explicit expressions of these operators are given in [13] and they will not be repeated here.

The fine-structure perturbation operators [14] are given by

$$H_{FS} = H_{SO} + H_{SOO} + H_{SS} \quad (10)$$

where the spin-orbit, the spin-other-orbit and the spin-spin operators, respectively, are

$$H_{SO} = \frac{Z\alpha^2}{2} \sum_{i=1}^4 \frac{\mathbf{l}_i \cdot \mathbf{s}_i}{r_i^3} \quad (11)$$

$$H_{SOO} = -\frac{\alpha^2}{2} \sum_{\substack{i,j=1 \\ i \neq j}}^4 \left[\frac{1}{r_{ij}^3} (\mathbf{r}_i - \mathbf{r}_j) \times \mathbf{P}_i \right] \cdot (\mathbf{s}_i + 2\mathbf{s}_j) \quad (12)$$

$$H_{SS} = \alpha^2 \sum_{\substack{i,j=1 \\ i < j}}^4 \frac{1}{r_{ij}^3} \left[\mathbf{s}_i \cdot \mathbf{s}_j - \frac{3(\mathbf{s}_i \cdot \mathbf{r}_{ij})(\mathbf{s}_j \cdot \mathbf{r}_{ij})}{r_{ij}^2} \right]. \quad (13)$$

The l_i and s_i stand for the orbital and the spin angular momentum of the i th electron and α is the fine structure constant. The wave function used to calculate these perturbations in the LSJ representation is

$$\Psi_{LSJJ_z} = \sum_{M, S_z} \langle LSMM_S | JM_J \rangle \psi_b(1, 2, 3, 4). \quad (14)$$

The fine-structure energy levels are calculated by the first-order perturbation theory

$$(\Delta E_{FS})_J = \langle \Psi_{LSJJ_z} | H_{SO} + H_{SOO} + H_{SS} | \Psi_{LSJJ_z} \rangle. \quad (15)$$

The hyperfine structure of atomic energy levels is caused by the interaction between the electrons and the electromagnetic multipole moments of the nucleus. The leading terms of this interaction are the magnetic dipole and electric quadrupole momentum. For an N -electron system, the hyperfine-interaction Hamiltonian can be represented as follows [16, 17]:

$$H_{hfs} = \sum_{k=1} T^{(k)} M^{(k)} \quad (16)$$

where $T^{(k)}$ and $M^{(k)}$ are spherical tensor operators of rank k in the electronic and nuclear space, respectively. The $k = 1$ term represents the magnetic-dipole interaction between the magnetic field generated by electrons and the nuclear magnetic dipole moments; the $k = 2$ term represents the electric quadrupole interaction between the

States	E_b	ΔE_{RV}	E_{nonrel}	ΔE_{rel}	E_{total}
$1s2s2p^2\ ^5P^e(1)$	-5.3865140	-0.0000897	-5.3866037	-0.0006007	-5.3872044
	-5.3865019 ^a				-5.3871726 ^a -5.386533 ^b
$1s2s2p3p\ ^5P^e(2)$	-5.3678751	-0.0000063	-5.3678814	-0.0006102	-5.3684916
$1s2s2p4p\ ^5P^e(3)$	-5.3675156	-0.0000064	-5.3675220	-0.0006097	-5.3681317
$1s2s2p5p\ ^5P^e(4)$	-5.3668659	-0.0000079	-5.3668738	-0.0006113	-5.3674851
$1s2p^3\ ^5S^o(1)$	-5.2560736	-0.0000337	-5.2561073	-0.0005564	-5.256637
	-5.2560529				-5.2566541 ^a -5.256047 ^b
$1s2p^23p\ ^5S^o(2)$	-5.2453331	-0.0000152	-5.2453483	-0.0005620	-5.2459103
$1s2p^24p\ ^5S^o(3)$	-5.2403270	-0.0000511	-5.2403781	-0.0005654	-5.2409435
$1s2p^25p\ ^5S^o(4)$	-5.2390759	-0.0001056	-5.2391815	-0.0005668	-5.2397483

^aReference [11], ^breference [2].

electric field gradient from electrons and the non-spherical charge distribution of the nucleus. The higher-order contribution terms are much smaller and can often be neglected.

In the non-relativistic framework, the electronic tensor operators in atomic units can be written as:

$$T^{(1)} = \frac{\alpha^2}{2} \sum_{i=1}^4 \left[2g_l r_i^{-3} l_i^{(1)} - \sqrt{10} g_s \left\{ s_i^{(1)} C_i^{(2)} \right\}^{(1)} r_i^{-3} + \frac{8\pi}{3} g_s s_i^{(1)} \delta(\mathbf{r}_i) \right] \quad (17)$$

and

$$T^{(2)} = - \sum_{i=1}^4 r_i^{-3} C_i^{(2)} \quad (18)$$

where $g_l = (1 - m_e/M)$ and $g_s = 2.0023193$ are the electron orbital and electron spin g factors, respectively. M is the nuclear mass. The tensor $C_i^{(2)}$ is connected with the spherical harmonics $Y_{lm}(i)$ by $C_m^{(l)} = \sqrt{4\pi/(2l+1)} Y_{lm}$.

The hyperfine interaction couples the electronic angular momenta \mathbf{J} and the nuclear angular momenta \mathbf{I} to a total angular momentum $\mathbf{F} = \mathbf{I} + \mathbf{J}$. The uncoupling and coupling hyperfine constants are defined in a.u. as [16, 17]:

$$a_C = \langle \gamma L S M_L M_S | \sum_{i=1}^N 8\pi \delta^3(\mathbf{r}_i) s_0(i) | \gamma L S M_L M_S \rangle \quad (\text{Fermi contact}) \quad (19)$$

$$a_{SD} = \langle \gamma L S M_L M_S | \sum_{i=1}^N 2C_0^{(2)}(i) s_0(i) r_i^{-3} | \gamma L S M_L M_S \rangle \quad (\text{spin-dipolar}) \quad (20)$$

$$a_l = \langle \gamma L S M_L M_S | \sum_{i=1}^N l_0(i) r_i^{-3} | \gamma L S M_L M_S \rangle \quad (\text{orbital}) \quad (21)$$

$$b_q = \langle \gamma L S M_L M_S | \sum_{i=1}^N 2C_0^{(2)}(i) r_i^{-3} | \gamma L S M_L M_S \rangle \quad (\text{electric quadrupole}) \quad (22)$$

Table 1. Energies (a.u.) of $1s2s2pnp\ ^5P$ ($n = 2-5$) and $1s2p^2mp\ ^5S$ ($m = 2-5$) for Li⁻ ion. E_b stands for the upper bound; ΔE_{RV} is the restricted variation correction; ΔE_{rel} is the relativistic correction and mass polarization.

and

$$A_J = \frac{\mu_I}{I} \frac{1}{[J(J+1)(2J+1)]^{1/2}} \langle \gamma J || T^{(1)} || \gamma J \rangle \quad (23)$$

$$B_J = 2Q \left[\frac{2J(2J-1)}{(2J+1)(2J+2)(2J+3)} \right]^{1/2} \langle \gamma J || T^{(2)} || \gamma J \rangle \quad (24)$$

where μ_I is the nuclear magnetic moment. Q is the nuclear electric quadrupole moment.

3 Result and discussion

In this work, two high- n series of Li⁻ ion, $1s2s2pnp\ ^5P$ and $1s2p^2mp\ ^5S^o$ states, with $n, m = 2-5$, are studied. The energies, the fine structure, and the hyperfine structure for these high- n systems are calculated using the saddle-point variational method and restricted variation method with accurate multiconfiguration-interaction wave functions constructed from Slater basis sets. The core-excited states of the Li⁻ ion are a complex four-electron atomic system. The correlation effects among electrons are complicated. Many relevant angular and spin couplings are important for the energy. In order to get a high-quality wave function, the number of angular-spin components in the wave functions ranges from 7 to 50, and the number of linear parameters ranges from 541 to 1298. In this work, for the $1s2s2pnp\ ^5P$ ($n = 2-5$) state, even parity, the important angular series $[l_1, l_2, l_3, l_4]$ is $[00l, l], [01l, (l+1)], [11l, l], [02(l+1), (l+1)], [11l, (l+2)], [02(l+1), (l+3)]$ etc. For the $1s2p^2mp\ ^5S^o$ ($m = 2-5$) state, odd parity, the available angular series $[l_1, l_2, l_3, l_4]$ is $[01l, l], [11l, (l+1)], [02(l+1), (l+2)], [12(l+1), (l+1)], [12(l+1), (l+3)], [22(l+1), (l+2)]$. In both cases, the value of l is from 1 to 9, as the energy contribution from the set with $l > 9$ is small and negligible. For each set of orbital angular momenta l_1, l_2, l_3 and l_4 , there could be several ways to couple this set into the desired total orbital angular momentum. In order to improve the energy E_b obtained from ψ_b , the restricted variational method is used to compute energy contributions from each chosen angular-spin series.

Table 2. Wavelength λ (nm), oscillator strength (f_l, f_v, f_a), and lifetime τ (ns) of the core-excited quintet states for Li^- .

Transitions	f_l	f_v	f_a	τ	λ
$1s2s2p^2\ ^5\text{P}^e \rightarrow 1s2p^3\ ^5\text{S}^o$	0.2104	0.2095	0.2236	2.89	349.06
	0.2095 ^a	0.2093 ^a	0.2277 ^a	2.91 ^a	349.12 ^a
				2.7 ^e	349.0 ^e
				2.3 ± 0.1^b	349.0 ^b
				2.86 ± 0.1^c	348.98 ^c
			2.9 ± 0.2^d	349.07 ^d	
$1s2s2p3p\ ^5\text{P}^e \rightarrow 1s2p^23p\ ^5\text{S}^o$	0.0550	0.0551	0.0505	12.56	371.72
$1s2s2p4p\ ^5\text{P}^e \rightarrow 1s2p^24p\ ^5\text{S}^o$	0.0582	0.0508	0.1465	11.02	358.26
$1s2s2p5p\ ^5\text{P}^e \rightarrow 1s2p^25p\ ^5\text{S}^o$	0.0632	0.0511	0.4193	10.05	356.72

^aReference [11], ^breference [5], ^creference [2], ^dreference [3], ^ereference [8].

Table 3. Fine structure splittings (in cm^{-1}) of the $1s2s2pnp\ ^5\text{P}$ ($n = 2-5$) states for Li^- .

States	$1s2s2p^2\ ^5\text{P}^e$		$1s2s2p3p\ ^5\text{P}^e$	$1s2s2p4p\ ^5\text{P}^e$	$1s2s2p5p\ ^5\text{P}^e$
	This work	Ref. [11]			
v_{2-1}	0.92	0.91	0.58	0.86	0.83
v_{3-2}	-1.32	-1.32	-0.64	-0.81	-0.84

Table 4. Hyperfine parameters of $1s2s2pnp\ ^5\text{P}$ ($n = 2-4$) and $1s2p^2mp\ ^5\text{S}$ ($m = 2-4$) states for Li^- (in a.u.).

Resonances	sources	a_c	a_l	a_{SD}	b_q
$1s2s2p^2\ ^5\text{P}^e(1)$	This work	111.1411	0.183978	0.03751	0.07502
	Others	37.059 ^a	0.1850 ^a	0.03833 ^a	
$1s2s2p3p\ ^5\text{P}^e(2)$	This work	112.2780	0.155001	0.03173	0.06345
$1s2s2p4p\ ^5\text{P}^e(3)$	This work	112.2656	0.155150	0.03168	0.06335
$1s2p^3\ ^5\text{S}^o(1)$	This work	106.0132			
	Others	35.366 ^a			
$1s2p^23p\ ^5\text{S}^o(2)$	This work	105.9522			
$1s2p^24p\ ^5\text{S}^o(3)$	This work	105.9558			

^aReference [8].

In Table 1, the energies of $1s2s2pnp\ ^5\text{P}$ ($n = 2-5$) and $1s2p^2mp\ ^5\text{S}^o$ ($m = 2-5$) states for Li^- ion are given, including the restricted energies and the relativistic energies. The high- n core-excited states are close to the ionization threshold of Li^- , and it is difficult to carry out high-quality theoretical calculations due to the numerical instability. The correlation effects between electrons in this system are very complicated. In order to get the high-quality wave function, all important angular-spin components have been included in the wave functions. Yang and Chung [11] first reported the lowest core-excited states $1s2s2p^2\ ^5\text{P}$ and $1s2p^3\ ^5\text{S}$ of Li^- . The results of the energies of $1s2s2p^2\ ^5\text{P}$ and $1s2p^3\ ^5\text{S}$ states in this work are lower than their theoretical data [11] about 32 and 10 $\mu\text{a.u.}$, and the difference with Bunge [2] is 671 and 617 $\mu\text{a.u.}$, respectively. To our knowledge, no calculations of these higher- n core-excited states have been reported. Our results should be useful in future experiments.

Table 2 gives the results of oscillator strengths from the dipole length (f_l), the dipole velocity (f_v), the dipole acceleration (f_a), the radiative lifetimes, and the transition wavelengths. For the low- n states, the three formulae (f_l, f_v, f_a) agree well; for the high- n core-excited states, because these oscillator strength are calculated with ψ_b

only, and the open-channel part of the wave function is not included, the three results agree reasonably well. The oscillator strength and the transition wavelength for Li^- in this work are in good agreement with existed data. In Table 2, the lifetime for $\text{Li}^- 1s2p^3\ ^5\text{S}$ state agrees well with those of Yang [11], Bunge [2], and Berry [3]. However, there is a discrepancy between the theoretical data and experimental result of Mannervik [5]. The result for the $^5\text{S}^o$ lifetime from Nicolaides [8], which includes the contribution of radiative autoionization, agrees with the experiment [5] better. The radiative autoionization might play an important role in this process. To our knowledge, no data of high- n states of this system have been reported.

In this work, the fine structure splittings are calculated with the H_{SO} , H_{SOO} and H_{SS} perturbation operators by using the first-order perturbation theory on the $1s2s2pnp\ ^5\text{P}$ ($n = 2-5$) states. The results are presented in Table 3. Our calculation results of the fine structure corrections are in agreement with those of Yang and Chung [11].

The hyperfine structure is caused by the interaction between the electrons and the electromagnetic multipole moments of the nucleus, sensitive to the correlation effects between electrons and the relativistic corrections. Tables 4 and 5 give the hyperfine parameters and the hyperfine

Table 5. Hyperfine coupling constants (in GHz) of $1s2s2pnp$ 5P ($n = 2-4$) and $1s2p^2mp$ 5S ($m = 2-4$) states for Li⁻.

Resonances	A_J			B_J		
	$J = 3$	$J = 2$	$J = 1$	$J = 3$	$J = 2$	$J = 1$
$1s2s2p^2$ $^5P^e(1)$	2.57614	3.19898	5.74867	6.52168(-4)	-6.52168(-4)	6.52168(-5)
	2.576 ^a	3.200	5.751 ^a			
	2.577 ^b	3.200 ^b	5.751 ^b			
$1s2s2p3p$ $^5P^e(2)$	2.59993	3.23202	5.80972	5.51649(-4)	-5.51649(-4)	5.51649(-5)
$1s2s2p4p$ $^5P^e(3)$	2.59965	3.23168	5.80905	5.50760(-4)	-5.50760(-4)	5.50760(-5)
$1s2p^3$ $^5S^o(1)$		3.66403				
		3.674 ^a				
		3.667 ^b				
$1s2p^23p$ $^5S^o(2)$		3.66192				
$1s2p^24p$ $^5S^o(3)$		3.66205				

^aReference [10], ^breference [8].

coupling constants for $1s2s2pnp$ 5P ($n = 2-4$) and $1s2p^2mp$ $^5S^o$ ($m = 2-4$) states in Li⁻. In this calculation, high precise wave functions are used. We studied the hyperfine structure parameters: Fermi contact a_c , the spin-dipolar a_{SD} , the orbital a_l , and the electric quadrupole b_q . In this work, $Q = 0.0370$, $\mu_I = 3.256427$ and $I = 3/2$ are taken from reference [19]. The hyperfine structures of $1s2p^3$ $^5S^o$ and $1s2s2p^2$ 5P states of Li⁻ ion are explored by Beck in 1983 [8]. Our results agree well with the data of Beck [8] and Yang [8].

4 Conclusion

In this work, the energies, the fine structure and the oscillator strengths of $1s2s2pnp$ 5P ($n = 2-4$) and $1s2p^2mp$ $^5S^o$ ($m = 2-4$) states of Li⁻ ion are calculated. The energies obtained in this work are much lower than the others previously published, the wavelengths and radiative lifetimes are in agreement with the published data. The hyperfine parameters and the hyperfine coupling constants are reported. The hyperfine interaction effect cannot be neglected for the low- n core-excited states. These results should be very useful for experiments in the future.

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