Energy levels and radiative lifetimes of $3pns\ ^3P_0$ and $3pnd\ ^3P_0$ series of Si I

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The energy levels and lifetimes of $3pns\ ^3P_0$ ($n = 7-35$) and $3pnd\ ^3P_0$ ($n = 6-17$) series of neutral silicon are calculated and predicted by means of multichannel quantum defect theory (MQDT). In addition, the perturbation caused by core-excited state $3s3p^3$ is discussed. The $3pnd\ ^3P_0$ series, especially $3p4d\ ^3P_0$, $3p5d\ ^3P_0$, and $3p6d\ ^3P_0$ are perturbed strongly by the core-excited state $3s3p^3\ ^3P_0$. These cause the lifetime of $3pnd\ ^3P_0$ ($n = 5-7$) to be less than that of $3p4d\ ^3P_0$. The lifetimes of $3p14d\ ^3P_0$ (65479.14 cm^{-1}) and $3p16d\ ^3P_0$ (65608.77 cm^{-1}) are less than that of their frontal states respectively, because these states are perturbed by $3p22s\ ^3P_0$ (65476.48 cm^{-1}) and $3p30s\ ^3P_0$ (65608.99 cm^{-1}) respectively.

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The knowledge of the radiative lifetime of excited atomic levels, especially Rydberg states of atoms, is particularly useful for understanding the atomic structure. The lifetime together with branching ratios provides information about transition probability and hence oscillator strength having important application in different branches of physics such as quantum electronics, atomic physics and spectroscopy, plasma physics, and astrophysics. The silicon atom is a semiconductor atom and is useful in many fields, contributing substantially to ultraviolet solar and stellar opacity through its high abundance and rich ultraviolet spectrum. The last experimental radiative lifetimes of Si I were measured by O'Brian *et al.*[1]. They reported the lifetimes of 47 upper levels in neutral silicon ranging in energy from 39000 to 63000 cm^{-1} measured by time-resolved laser-induced fluorescence on a slow atomic beam of silicon using radiative transitions in the visible, ultraviolet, and vacuum-ultraviolet (VUV) regions between 160 and 410 nm. For the lifetimes of odd-parity $J = 0$ of neutral silicon, only $3pns\ ^3P_0$ ($n = 3-5$) and $3pns\ ^3P_0$ ($n = 4-6$) were determined. In 2002, Coutinho *et al.* calculated the weighted oscillator strengths and lifetimes for the Si I spectrum by means of multiconfiguration Hartree-Fock relativistic approach[2], but some lifetimes of levels disagreed with experiment, and some lifetimes of levels were not calculated. In this letter, the energy values and lifetimes of Rydberg series $3pns\ ^3P_0$ and $3pnd\ ^3P_0$ are calculated using the multichannel quantum defect theory (MQDT). Energy levels and lifetime values for highly excited Rydberg states are predicted and the perturbation caused by $3s3p^3$ is discussed.

MQDT has been evolved into a powerful tool for the interpretation of highly excited Rydberg structure of atoms and ions[3−5]. The main idea of MQDT is taking the atom or ion as a system of one excited electron and the ion core. The interaction between the excited electron and the ion core is separated into two regions: $r > r_0$ and $r < r_0$. The wave function of $r > r_0$ region is a combination of Coulomb wave functions and the wave function of $r < r_0$ region is described by an eigenquantum defects μ_α and an orthogonal transformation matrix $U_{i\alpha}$. μ_α and $U_{i\alpha}$ are essential MQDT parameters[6,7] determined by fitting experimental data on energy levels. To ensure correct asymptotic behavior of the wave functions for discrete levels, the following condition is required:

$$\det |U_{i\alpha} \sin \pi(\nu_i + \mu_\alpha)| = 0, \tag{1}$$

where ν_i is the effective quantum number corresponding to the ith ionization limit I_i. The discrete energy level E of an excited atom is given by

$$E = I_i - \frac{R}{\nu_i^2}, \quad (i = 1, 2, \cdots, N) \tag{2}$$

where R is the mass-connected Rydberg constant, N is the total number of dissociation channels.

The MQDT normalized wave function for a highly excited state ψ_n at $r > r_0$ is represented as a superposition of the dissociation channels (the ith channel) as

$$\psi_n = \sum_i \phi_i p_i^{(n)} z_i^{(n)}, \tag{3}$$

where $p_i^{(n)}$ is the electron wave function and can be expressed in terms of the analytically known Whittaker function, ϕ_i denotes the wave function of the residual ion core, the spin, and the angular momentum coupling in the ith channel. The admixture coefficient of the wave function of a discrete energy level E_n in the ith channel is

$$z_i^{(n)} = (-1)^{l_i+1} (\nu_{i,n})^{3/2} \sum_\alpha U_{i\alpha} \cos \pi(\nu_{i,n} + \mu_\alpha) A_\alpha^{(n)} / N_n, \tag{4}$$

where $A_\alpha^{(n)}$ is the coefficient of the eigenchannel expansion, N_n is a normalization factor, and l_i is the angular momentum quantum number of excited electron in the ith channel. According to Refs. [8,9], the radiative decay rate of the nth level can be written as

$$\Gamma_n = \sum_i \sum_j z_i^{(n)} z_j^{(n)} (\nu_i^{(n)} \nu_j^{(n)})^p \Gamma_{ij}, \tag{5}$$

where Γ_{ij} no longer depends on energy but on channel, p is an adjustable parameter.

Finally, we obtain the natural lifetime of the atom in the nth excited state as

$$\tau_n = \frac{1}{\Gamma_n}. \tag{6}$$

The parameters p and Γ_{ij} in Eq. (5) can be determined by fitting experimental data of available lifetimes.

According to the recent data on Si I[10], only configuration $3s2p^3\ ^3P_0$ lies under the first ionization limit, so we select $3s3p^2np\ ^3P_0$ as the perturbing states of $3pns\ ^3P_0$ and $3pnd\ ^3P_0$ channels. The MQDT parameters of Si I for a set of $J = 0$ odd-parity model with three ionization limits and three channels are listed in Table 1. The energies are represented by effective quantum number in MQDT ($E \propto 1/\nu^2$) and μ_α is energy-dependent. In order to calculate the energy accurately, μ_α which is the function of high power of energy, can be considered to be $\mu_\alpha^0 + \frac{1}{\nu^2}\mu_\alpha^1 + \frac{1}{\nu^4}\mu_\alpha^2$ and the admixture angles θ_{ij} (see Ref. [7]) no longer depend on energy. Using experimental energy levels from 39683.163 to 64859.23 cm^{-1}given by Ref. [10] to minimize $w = \sum_n \left[\frac{(\det|U_{i\alpha}\sin\pi(\nu_{i\alpha}+\mu_\alpha)|^2}{N_t}\right]^{1/2}$, we obtain the optimal MQDT parameters, which are listed in Table 1. In addition, the admixture coefficient of each channel is calculated using Eq. (4). By fitting experimental lifetimes given in Ref. [1], we obtain the parameters of p and Γ_{ij}, which are listed in Table 2. Figure 1 shows the admixture coefficients Z_1^2, Z_2^2, and Z_3^2 of $3pnd\ ^3P_0$ channel as functions of the principal quantum number n.

Table 1. MQDT Parameters of Si I for Odd-Parity $J = 0$

i	1	2	3
$\|\alpha\rangle$	$(p_{3/2}, p_{3/2})_0$	$(^2D_{3/2}, p_{3/2})_0$	$(p_{1/2}, s_{1/2})_0$
$\|i\rangle$	$3pnd\ ^3P_0$	$3s2p^2np\ ^3P_0$	$3pns\ ^3P_0$
I_i (cm^{-1})	66035.00	121057.11	65747.76
μ_α^0	0.1098	0.6236	0.8857
μ_α^1	−0.6787	0.6032	0.2267
μ_α^2	−7.2152	1.8847	0.1412
θ_{ij}	$\theta_{12} = -0.233$	$\theta_{13} = 0.095$	$\theta_{23} = 0.026$
$U_{i\alpha}$	−0.969	−0.228	0.098
	−0.230	0.973	−0.004
	0.094	0.026	0.995

Table 2. Values of Γ ($\Gamma_{ij} = \Gamma_{ji}$) and p

ns Series ($p = -3.2899$)			
i/j	1	2	3
1	0.28014×10^3		
2	0.17809×10^6	-0.52532×10^2	
3	-0.66985×10^5	-0.24913×10^3	0.28071×10^1
nd Series ($p = -3.450$)			
i/j	1	2	3
1	0.34156×10^2		
2	-0.26087×10^2	0.23459×10^4	
3	0.57740×10^1	-0.10517×10^4	-0.41514×10^5

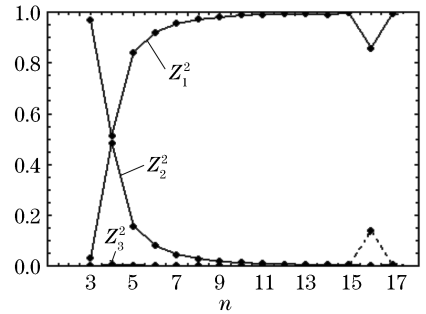

Fig. 1. Admixture coefficients Z_1^2, Z_2^2, and Z_3^2 of $3pnd\ ^3P_0$ channel as functions of the principal quantum number n.

Using MQDT parameters in Table 1, we calculate the energy levels of highly excited states (listed in Table 3) and admixture coefficients. Energy values of $3pns\ ^3P_0$ ($n = 22 - 35$) are predicted. Using lifetime parameters, we calculate the lifetimes of $3pns\ ^3P_0$ ($n = 7 - 35$) and $3pnd\ ^3P_0$ ($n = 6 - 17$) of neutral silicon, as listed in Tables 3 and 4.

The results in Tables 3 and 4 show that the agreement between theoretical and experimental energies is quite satisfactory. Table 1 shows that the admixture angle $\theta_{12} = -0.233$ between channel 1 and channel 2 is large, which indicates that the $3pnd\ ^3P_0$ series are perturbed strongly by the core-excited state $3s3p^3\ ^3P_0$, especially $3p4d\ ^3P_0$, $3p5d\ ^3P_0$, and $3p6d\ ^3P_0$ (see Fig. 1). The energy of core-excited $3s3p^3\ ^3P_0$ calculated in Ref. [2] is 56733.368 cm^{-1}, but there is no corresponding experiment value in Ref. [10], and the energy of $3p4d\ ^3P_0$ given in Ref. [10] is 56733.38 cm^{-1}. In this work, there is an energy of 56733.07 cm^{-1}, and the admixture coefficients of this energy in three channels are 0.511, 0.484, and 0.005, respectively. So we consider 56733.07 cm^{-1} as the state $3p4d\ ^3P_0$. However, we have not obtained the new energy of $3s3p^3\ ^3P_0$. The reason is that the core-excited state $3s3p^3\ ^3P_0$ has been mixed by $3pnd\ ^3P_0$ series, but

Table 3. Energy Levels and Lifetimes of $3pnd\ ^3P_0$ for Si I Calculated by MQDT

	Energy Level (cm^{-1})		Lifetime (ns)		
n	This Work	E_{exp}[9]	This Work	Theory[2]	τ_{exp}[1]
3	50602.94	50602.44	11.5	3.121	11.5 ± 0.6
4	56733.07	56733.38	31.7	6.062	31.9 ± 1.6
5	61962.50	61960.26	12.4	12.4	12.1 ± 0.6
6	63123.11	63123.36	18.8	20.55	
7	63863.30	63863.80	28.7		
8	64358.12	64358.52	42.9	47.62	
9	64703.13	64703.23	61.5	65.06	
10	64952.49	64952.58	86.9	93.45	
11	65138.25	65138.22	118.0	110.0	
12	65280.19	65280.10	153.6	141.7	
13	65391.02	65390.91	204.8	176.9	
14	65479.12	65479.14	205.7	214.1	
15	65550.44	65550.33	325.1	318.7	
16	65608.77	65608.63	276.7	359.2	
17	65657.31	65657.24	435.9	345.5	

Table 4. Energy Levels and Lifetimes of $3pns\ ^3P_0$ for Si I Calculated by MQDT

n	Energy Level (cm^{-1}) This Work	E_{exp}[9]	Lifetime (ns) This Work	Theory[2]	τ_{exp}[1]
4	39683.14	39683.16	4.5	4.478	4.5 ± 0.2
5	54245.96	54245.02	13.9	6.889	13.8 ± 0.7
6	59221.57	59221.11	32.6	32.990	32.7 ± 1.6
7	61538.43	61538.05	71.3	64.51	
8	62806.78	62806.65	118.5	113.20	
9	63576.88	63576.85	170.9	134.70	
10	64079.59	64079.74	248.9	205.5	
11	64425.86	64425.97	354.6	295.8	
12	64674.49	64674.61	473.4	409.7	
13	64859.03	64859.23	623.0	551.3	
14	64999.76	65000.11	788.8		
15	65109.51	65109.47	942.3	911.8	
16	65196.78	65196.82	1132.9	1154.0	
17	65267.28	65267.35	1299.4	1549.0	
18	65325.08	65324.68	1505.7		
19	65373.04	65372.98	1683.8	1706.0	
20	65413.28	65413.38	1892.2	1987.0	
21	65447.36	65447.45	2068.2	4569.0	
22	65476.48		2292.9		
35	65647.68		4152.6		

its energy level should be near 56733.07 cm^{-1}. There is a state with the energy of 60052.79 cm^{-1} and the admixture coefficients of this energy in three channels are 0.677, 0.318, and 0.005. This energy is the state of $3pnd\ a^3P_0$, which is 60042.5 cm^{-1} in Ref. [10].

The calculated lifetimes of $3pnd\ ^3P_0$ in Table 3 are in good agreement with the results of Ref. [2]. Table 3 indicates that the lifetime of $3p5d\ ^3P_0$ is less than that of $3p4d\ ^3P_0$, because the perturbation comes from the state of $3s3p^3\ ^3P_0$. Although it can be seen from Fig. 1 that although $3p4d\ ^3P_0$ is the most perturbed state, its lifetime does not show diminishing as a result of using the lifetime of $3p4d\ ^3P_0$ in fitting experimental data. The lifetimes of $3p14d\ ^3P_0$ (65479.14 cm^{-1}) and $3p16d\ ^3P_0$ (65608.77 cm^{-1}) are less than that of their frontal states respectively, because these states are perturbed by $3p22s\ ^3P_0$ (65476.48 cm^{-1}) and $3p30s\ ^3P_0$ (65608.99 cm^{-1}) respectively. Theoretically, this is the result of stronger interaction between a couple of interaction configurations with too small energy interval. It can be seen from Ref. [2] that the lifetime of $3p17d\ ^3P_0$ is less than that of $3p16d\ ^3P_0$, but the result of this work indicates that $3p17d\ ^3P_0$ is not perturbed by other energy states. According to the MQDT, this is an impossible emergence, so it should be accurate that the lifetime value of $3p17d\ ^3P_0$ calculated by this work is larger than that of Ref. [2].

Because the perturbation of $3pns\ ^3P_0$ series coming from the other two channels is very small, the energy states of this series in their own channel compositions are more than 98% and only individual energy state is perturbed slightly. So the figure of admixture coefficients is not given. Table 4 shows that the calculated results agree well with Ref. [2], but the calculated lifetime value of $3p21s\ ^3P_0$ is less than half of that in Ref. [2]. Generally, without perturbation, the lifetime of a Rydberg series meets the formula $\tau = \tau_0 \nu^{\alpha}$ approximatively, where τ_0 and α are constants and ν is the effective quantum number. The lifetime value of $3p21s\ ^3P_0$ calculated by this formula ($\tau = 3.885\nu^{2.1395}$ fitted by using lifetimes of $3pns\ ^3P_0$ ($n = 15 - 20$)) is about 2141.6 ns, which is very close to this work (2068.2 ns). In addition, the lifetime value of $3p30s\ ^3P_0$ (energy level 65608.99 cm^{-1}) is less than that of $3p29s\ ^3P_0$ due to the small perturbation coming from $3p16d\ ^3P_0$ (energy level 65608.63 cm^{-1}).

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References

1. T. R. O'Brian and J. E. Lawler, Phys. Rev. A **44,** 7134 (1991).
2. L. H. Coutinho and A. G. Trigueiros, J. Quant. Spectrosc. Radiat. Transf. **75,** 357 (2002).
3. K. T. Lu, Phys. Rev. A **4,** 579 (1971).
4. J. P. Connerade, *Highly Excited Atoms* (Cambridge University Press, New York, 1998).
5. C. Zhou, L. Liang, and L. Zhang, Chin. Opt. Lett. **5,** 438 (2007).
6. U. Fano and R. Racah, *Irreducible Tensorial Sets* (Academic, New York, 1959) p.68.
7. C.-M. Lee and K. T. Lu, Phys. Rev. A **8,** 1241 (1973).
8. X.-W. Liu and Z.-W. Wang, Phys. Rev. A **40,** 1838 (1989).
9. L. Liang and Y. C. Wang, J. Phys. B **36,** 4387 (2003).
10. Yu. Ralchenko, A. E. Kramida, J. Reader, and NIST ASD Team, NIST Atomic Spectra Database Levels Data, http://physics.nist.gov/cgi-bin/ASD/energy1.pl (May 10, 2008).