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Calculations of radiative lifetimes of Rydberg $6pnd\ J=2$ states of Pb I by multichannel quantum defect theory

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Recently measured radiative lifetimes of the Rydberg series $6pnd\ ^1[\frac{5}{2}]^o_2$ and $^1[\frac{3}{2}]^o_2$ ($n=6-13$) of Pb I [Li *et al.*, Phys. Rev. A **57**, 3443 (1998)] have been compared with theoretical values obtained by means of multichannel quantum defect theory (MQDT) analyses. The MQDT parameters were obtained from experimentally determined energy levels. The channel admixture coefficients were calculated and used to evaluate the theoretical lifetimes. In addition, lifetime values for highly excited Rydberg states have been predicted.

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Multichannel quantum defect theory (MQDT) has been applied extensively to the description of perturbed Rydberg series. Previously, the study of Rydberg series focused on atoms possessing Rydberg states composed of an *s*-electron ion core and a highly excited electron [1–3]. However, in the present study of neutral lead, the Rydberg states consist of a *p*-electron ion core and a highly excited electron; the ion core ($6s^26p$) splits into the levels $^2P^o_{1/2}$ (lower ionization limit) and $^2P^o_{3/2}$ (upper ionization limit). In addition, the orbital-spin coupling in a lead atom is strong enough that the ground configuration ($6p^2$) and excited configurations ($6pns$ and $6pnp$) are adequately described by *jj* coupling, while the configurations with high-angular-momentum electrons ($6pnd$,$6pnf$) are close to *jK* coupling. The high-lying states of the carbon group also offer a chance to understand the chemical reactions of excited atomic components [4]. For the reasons mentioned above, it is of interest to investigate the Rydberg states of this kind of atom not only in theory but also in practice.

The energy positions of Pb I Rydberg states have been determined using conventional absorption spectroscopy [5] and laser spectroscopy [4,6–8]. In order to give more insight into the properties of Rydberg states of Pb I, additional physical quantities such as radiative lifetimes are required. Since the resonant lines of Pb I lie in the uv and vacuum uv regions, selective excitation and detection are rather difficult, and only a limited number of experimental lifetimes of lower energy levels have been measured using different techniques [9–11]. To make a systematic investigation for Pb Rydberg states, we recently performed lifetime measurements on the Rydberg states of Pb I by means of time-resolved laser spectroscopy and laser ablation techniques [12]. In this paper, we use a MQDT analysis of our measured lifetimes of Pb I for deeper understanding of the Rydberg-state features of this kind of atom and to test MQDT theory.

In the Rydberg formula, the discrete energy level E of an excited atom is given by

$$E=I_i-\frac{R}{\nu_i^2}=I_j-\frac{R}{\nu_j^2} \tag{1}$$

where I_i and ν_i are the ith ionization limit and corresponding effective quantum number, respectively, and R is the mass-corrected Rydberg constant [13].

The interaction between the excited electron and the ion core is separated into two regions: (I) $r>r_0$ (r_0 is the radius of the ion core) and (II) $r<r_0$. The interaction in region I is Coulombic and is described by the collision channels (i channel) identified in *jj* coupling, while the interaction in region II is non-Coulombic and is described by the eigenchannels (α channel). The two kinds of channel are connected by an orthogonal transformation matrix $U_{i\alpha}$. To ensure correct asymptotic behavior of the wave functions for discrete levels the following condition is required:

$$\det\left|U_{i\alpha}\sin\pi(\nu_i+\mu_\alpha)\right|=0, \tag{2}$$

where μ_α is the eigenquantum defect of eigenchannel α.

For a Rydberg series of configuration involving M interacting channels and N relevant series limits, there are $N-1$ independent equations similar to Eq. (1), and each equation determines a line L in the N-dimensional space of ν_i. Equation (2) describes a surface S in the same space. Because each bound state simultaneously satisfies Eqs. (1) and (2), the state energy can be calculated from the intersections of L and S. μ_α and $U_{i\alpha}$ are essential MQDT parameters. In MQDT analyses, the theoretical energy levels are fitted to the experimental ones by adjusting the parameters μ_α and $U_{i\alpha}$. It is very convenient to introduce an intermediate basis of pure coupled channels $\overline{\alpha}$, and express the matrix $U_{i\alpha}$ as

$$U_{i\alpha}=\sum_{\overline{\alpha}} U_{i\overline{\alpha}}V_{\overline{\alpha}\alpha}, \tag{3}$$

where $U_{i\overline{\alpha}}$ is the transformation matrix between channels i and $\overline{\alpha}$ and can be obtained analytically from the 6-j symbols [14]; $V_{\overline{\alpha}\alpha}$ is an orthogonal matrix generated by $M(M-1)/2$ successive rotations θ_{ij} [15]. Thus the angles θ_{ij} become MQDT parameters instead of $U_{i\alpha}$.

TABLE I. MQDT parameters for the odd-parity $J=2$ bound levels of Pb I.

$I,\overline{\alpha},\alpha$	1	2	3	4
$\|i\rangle$	$6pnd\ (3/2)[5/2]^o_2$	$6pnd\ (3/2)[3/2]^o_2$	$6pnd\ (1/2)[5/2]^o_2$	$6pnd\ (1/2)[3/2]^o_2$
I_i	73 900.64	73 900.64	59 819.57	59 819.57
$\|\overline{\alpha}\rangle$	$(3/2,5/2)^o_2$	$(3/2,3/2)^o_2$	$(1/2,5/2)^o_2$	$(1/2,3/2)^o_2$
μ^0_α	0.325	0.29	0.28	0.19
μ^1_α	−0.2539	0.2964	−0.4878	0.0330
$V_{\overline{\alpha}\alpha}$	$\theta_{13}=0.1787$	$\theta_{14}=0.6890$	$\theta_{23}=0.0560$	$\theta_{24}=0.2249$
$U_{i\alpha}$	0.3038	0.8323	−0.1223	−0.4472
	0.6961	−0.5261	−0.1403	−0.4679
	0.6504	0.1744	0.1965	0.7127
	0.0073	−0.0065	0.9627	−0.2705

The MQDT wave function Ψ_n at the nth, level can be expressed by the expansion of the wave functions Ψ_i of collision channels, i.e.,

$$\Psi_n=\sum_i Z_i^{(n)}\Psi_i \tag{4}$$

where $Z_i^{(n)}$ is the admixture coefficient of the i channel and is given by

$$Z_i^{(n)}=(-1)^{l_i+1}(\nu_i^{(n)})^{3/2}\sum_\alpha U_{i\alpha}\cos(\nu_i^{(n)}+\mu_\alpha)A_\alpha^{(n)}/N_n, \tag{5}$$

where $A_\alpha^{(n)}$ is the coefficient of eigenchannel expansion and N_n is a normalization factor.

Utilizing MQDT wave functions, the lifetimes of Rydberg levels can be calculated by using parametric theory [16]. Because M interacting channels consist of perturbing channels and perturbed channels, from Eq. (4) the wave function is rewritten as

$$\Psi_n=\sum_i Z_i^{(n)}\Psi_i+\sum_\alpha Z_\alpha^{(n)}\Psi_\alpha \tag{6}$$

where Ψ_i and Ψ_α are the perturbed and perturbing channel wave functions, respectively. In the electronic dipole approximation, the radiative decay rate Γ_n of the level n can be given by

$$\Gamma_n=\sum_i (Z_i^{(n)})^2\Gamma_i+\sum_\alpha (Z_\alpha^{(n)})^2\Gamma_\alpha, \tag{7}$$

where Γ_i and Γ_α are the decay rates of pure Rydberg levels, which are proportional to $1/(\nu^{(n)})^3$. Thus Eq. (7) becomes

$$\Gamma_n=\sum_i (Z_i^{(n)})^2\frac{\gamma_i}{(\nu_i^{(n)})^3}+\sum_\alpha (Z_\alpha^{(n)})^2\frac{\gamma_\alpha}{(\nu_\alpha^{(n)})^3}, \tag{8}$$

where γ_i and γ_α are lifetime parameters not dependent on the level n. In general, as the coefficients $Z_\alpha^{(n)}$ have considerable values only for the levels near the perturbers, the variation of Γ_n with $\nu_\alpha^{(n)}$ can be neglected, and Γ_n reduces to

$$\Gamma_n=\sum_i (Z_i^{(n)})^2\frac{\gamma_i}{(\nu_i^{(n)})^3}+\sum_\alpha (Z_\alpha^{(n)})^2\Gamma_\alpha, \tag{9}$$

TABLE II. The admixture coefficients $(Z_i^{(n)})^2$ for the odd-parity $J=2$ levels of $6pnd$ $(n=6-13)$ configurations.

Level	E_{expt} [5] (cm^{-1})	$(Z_i^{(n)})^2$ (units of 0.01) $(3/2)[5/2]^o_2$	$(3/2)[3/2]^o_2$	$(1/2)[5/2]^o_2$	$(1/2)[5/2]^o_2$
$6p6d\ (1/2)[5/2]^o_2$	45 443.17	0.9118	0.2836	96.2915	2.0131
$6p6d\ (1/2)[3/2]^o_2$	46 060.84	0.0547	0.0601	1.7392	94.1458
$6p7d\ (1/2)[5/2]^o_2$	52 101.66	0.3033	0.0958	99.0269	0.2377
$6p7d\ (1/2)[3/2]^o_2$	52 311.32	0.0013	0.0026	0.2033	98.1684
$6p8d\ (1/2)[5/2]^o_2$	55 003.29	0.2705	0.0888	99.4275	0.0036
$6p8d\ (1/2)[3/2]^o_2$	55 084.14	0.0002	0.0001	0.0057	99.1919
$6p9d\ (1/2)[5/2]^o_2$	56 526.49	0.3224	0.0949	99.3300	0.1156
$6p9d\ (1/2)[3/2]^o_2$	56 563.20	0.0020	0.0001	0.1211	99.4236
$6p10d\ (1/2)[5/2]^o_2$	57 424.02	0.4539	0.0956	99.1708	0.1878
$6p10d\ (1/2)[3/2]^o_2$	57 444.52	0.0048	0.0002	0.1890	99.5238
$6p11d\ (1/2)[5/2]^o_2$	57 995.90	0.8037	0.0606	98.9280	0.1529
$6p11d\ (1/2)[3/2]^o_2$	58 012.05	0.0100	0.0000	0.1500	99.6515
$6p12d\ (1/2)[5/2]^o_2$	58 378.56	3.5398	0.1566	96.3637	0.0223
$6p12d\ (1/2)[3/2]^o_2$	58 398.71	0.0445	0.0085	0.0121	99.8030
$6p6d\ (3/2)[5/2]^o_2$	58 517.71	59.9434	37.2720	5.5282	0.0611
$6p13d\ (1/2)[5/2]^o_2$	58 666.94	0.1616	2.4771	96.9876	0.3043

where Γ_α is independent of n. The lifetime τ_n of the level n can be calculated from the radiative decay rate Γ_n as $\tau_n = 1/\Gamma_n$. The parameters γ_i and Γ_α can be determined by fitting theoretical lifetimes to experimental lifetimes with the admixture coefficients obtained from MQDT analyses.

The odd-parity $J=2$ Rydberg series $6pnd$ of Pb I has four interacting channels: $6pnd\ \frac{1}{2}[\frac{5}{2}]^o_2$ and $6pnd\ \frac{1}{2}[\frac{3}{2}]^o_2$ converge to the first ionization limit $^2P_{1/2}$ (59 819.57 cm^{-1}) and $6pnd\ \frac{3}{2}[\frac{5}{2}]^o_2$ and $6pnd\ \frac{3}{2}[\frac{3}{2}]^o_2$ to the second one $^2P_{3/2}$ (73 900.64 cm^{-1}). Using experimental energy levels from 45 443.17 to 59 787.87 cm^{-1} reported by Brown, Tilford, and Ginter [5], we made MQDT calculations in a four-channel and two-limit model. μ_α and θ_{ij} are slowly varying functions of energy. Here the energy dependence of μ_α is considered to be $\mu_\alpha = \mu^0_\alpha + \varepsilon\mu^1_\alpha$, where $\varepsilon = (I_i - E)/R$. The energy dependence of θ_{ij} is neglected, as is done in general. Although $V_{\overline{\alpha}\alpha}$ depends on six angles θ_{ij}, the levels studied here can be fitted very well by setting $\theta_{12} = \theta_{34} = 0$ and adjusting other angles. The optimal MQDT parameters obtained by the nonlinear minimization method are listed in Table I.

To calculate the lifetimes of the odd-parity $6pnd$ ($n = 6-13$) $J=2$ levels measured by us [12], $(Z_i^{(n)})^2$ is calculated for these levels from Eq. (5) and the MQDT parameters and listed in Table II. From Eq. (9), the formula for radiative decay rate for these levels is given by

$$\Gamma_n = (Z^{(n)}_{(1/2)(5/2)^o_2})^2 \frac{\gamma_1}{(\nu_1^{(n)})^3} + (Z^{(n)}_{(1/2)[3/2]^o_2})^2 \frac{\gamma_2}{(\nu_1^{(n)})^3} + (Z^{(n)}_{(3/2)5/2]^o_2})^2 \Gamma_{(3/2)[5/2]^o_2} + (Z^{(n)}_{3/2[3/2]^o_2})^2 \Gamma_{(3/2)[3/2]^o_2}, \quad (10)$$

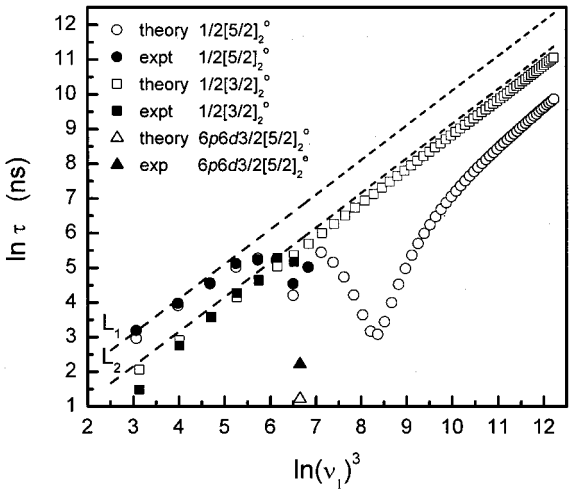

FIG. 1. Comparison of the theoretical and experimental lifetimes for the $6pnd$ $(1/2)[5/2]^o_2$ and $6pnd$ $(1/2)[3/2]^o_2$ states. The straight dashed lines L_1 and L_2 correspond to $\tau = (\nu_1)^3/\gamma_1$ and $(\nu_1)^3/\gamma_2$, respectively.

where γ_1 and γ_2 are parameters of the $6pnd\ \frac{1}{2}[\frac{5}{2}]^o_2$ and $\frac{1}{2}[\frac{3}{2}]^o_2$ channels, ν_1 is the effective quantum number in the first limit and $\Gamma_{(3/2)[5/2]^o_2}$ and $\Gamma_{(3/2)[3/2]^o_2}$ are parameters of the $6pnd\ \frac{3}{2}[\frac{5}{2}]^o_2$ and $\frac{3}{2}[\frac{3}{2}]^o_2$ channels. Using the nonlinear minimization method, we fitted the theoretical lifetimes to the experimental values with the MQDT admixture coefficients given in Table II and obtained the lifetime parameters γ_1

TABLE III. Lifetimes calculated by MQDT, experimental lifetimes, and comparison with previous results calculated by RHF theory for the $6pnd$ ($n=6-13$) series, and calculated lifetimes of the $6pnd$ $(1/2)[5/2]^o_2$ ($n=14-33$) series.

Level	E_{expt} [5] (cm^{-1})	Lifetime (ns) Calculated by MQDT	Expt. [12]	Calculated by RHF [12]	Level[a]	E_{expt} [5] (cm^{-1})	Calculated lifetime (ns)
$6p6d$ $(1/2)[5/2]^o_2$	45 443.15	19.40	24.5	24.75	$6p14d$ $(1/2)[5/2]^o_2$	58 865.40	233.3
$6p6d$ $(1/2)[3/2]^o_2$	46 060.84	7.87	4.4	3.81	$6p15d$ $(1/2)[5/2]^o_2$	59 017.08	174.6
$6p7d$ $(1/2)[5/2]^o_2$	52 101.66	50.30	53.7	66.44	$6p16d$ $(1/2)[5/2]^o_2$	59 134.25	114.4
$6p7d$ $(1/2)[3/2]^o_2$	52 311.32	18.41	15.7	24.24	$6p17d$ $(1/2)[5/2]^o_2$	59 226.10	68.3
$6p8d$ $(1/2)[5/2]^o_2$	55 003.29	96.67	94.5	121.9	$6p18d$ $(1/2)[5/2]^o_2$	59 298.63	38.5
$6p8d$ $(1/2)[3/2]^o_2$	55 084.14	36.40	35.4	88.02	$6p19d$ $(1/2)[5/2]^o_2$	59 356.35	23.9
$6p9d$ $(1/2)[5/2]^o_2$	56 526.49	151.95	169.0	195.9	$6p20d$ $(1/2)[5/2]^o_2$	59 403.89	21.9
$6p9d$ $(1/2)[3/2]^o_2$	56 563.20	63.79	72.0	30.94	$6p21d$ $(1/2)[5/2]^o_2$	59 445.18	31.5
$6p10d$ $(1/2)[5/2]^o_2$	57 424.02	196.39	187.0	324.4	$6p22d$ $(1/2)[5/2]^o_2$	59 482.07	54.6
$6p10d$ $(1/2)[3/2]^o_2$	57 444.52	102.38	105.0	94.79	$6p23d$ $(1/2)[5/2]^o_2$	59 514.75	92.6
$6p11d$ $(1/2)[5/2]^o_2$	57 995.90	191.75	182.0	411.4	$6p24d$ $(1/2)[5/2]^o_2$	59 543.50	146.0
$6p11d$ $(1/2)[3/2]^o_2$	58 012.05	153.66	199.0	201.7	$6p25d$ $(1/2)[5/2]^o_2$	59 568.60	214.9
$6p12d$ $(1/2)[5/2]^o_2$	58 378.56	67.55	94.2	127.3	$6p26d$ $(1/2)[5/2]^o_2$	59 590.56	299.7
$6p12d$ $(1/2)[3/2]^o_2$	58 398.71	213.07	176.0	496.1	$6p27d$ $(1/2)[5/2]^o_2$	59 609.90	400.7
$6p6d$ $(3/2)[5/2]^o_2$	58 517.71	3.38	9.2	4.24	$6p28d$ $(1/2)[5/2]^o_2$	59 626.94	518.0
$6p13d$ $(1/2)[5/2]^o_2$	58 666.94	151.52	152.0	206.5	$6p29d$ $(1/2)[5/2]^o_2$	59 641.90	652.0
					$6p30d$ $(1/2)[5/2]^o_2$	59 655.19	803.6
					$6p31d$ $(1/2)[5/2]^o_2$	59 667.07	972.6
					$6p32d$ $(1/2)[5/2]^o_2$	59 677.73	1160.1
					$6p33d$ $(1/2)[5/2]^o_2$	59 687.36	1366.7

[a]Designations determined from the admixture coefficient.

$=9.947\times10^8\ s^{-1}$, $\gamma_2=30.702\times10^8\ s^{-1}$, $\Gamma_{(3/2)[5/2]_2^o}=3.686\times10^8\ s^{-1}$, and $\Gamma_{3/2[3/2]_2^o}=1.998\times10^8\ s^{-1}$. Comparisons between the calculated and measured lifetimes of the $6pnd\ \frac{1}{2}[\frac{5}{2}]_2^o$ and $\frac{1}{2}[\frac{3}{2}]_2^o$ channels are shown in Fig. 1. The very satisfactory agreement seen in Fig. 1 indicates that MQDT parameters and admixture coefficients obtained in this work are reliable for describing the properties of Rydberg states. To compare these with previous theoretical results using the relativistic Hartree-Fock (RHF) method [12], the two kinds of theoretical result are listed in Table III together with experimental data. It is found that the theoretical lifetimes calculated by MQDT are more consistent with the measured values than those calculated by RHF theory.

Using the lifetime parameters mentioned above, lifetime calculations for the odd-parity $6pnd\ J=2$ levels are extended to the level 59 787.87 cm^{-1} and 116 lifetimes are obtained. The theoretical lifetimes with no corresponding experimental results are also given in Fig. 1, which shows that there are decreases of the lifetime in two regions of ν_1. The decrease at smaller ν_1 is due to strong perturbation of the perturbing channel $6p6d\ \frac{3}{2}[\frac{5}{2}]_2^o$ (58 517.71 cm^{-1}). The decrease at larger ν_1 occurs around the 59 403.89 cm^{-1} level where large channel admixtures occur. The admixture coefficients $(Z_i^{(n)})^2$ of the perturbing channels $\frac{3}{2}[\frac{5}{2}]_2^o$ and $\frac{3}{2}[\frac{3}{2}]_2^o$ are shown in Fig. 2. Only a very small admixture influences the $\frac{1}{2}[\frac{3}{2}]_2^o$ channel; the level lifetimes in this channel essentially retain the character of $(\nu_1)^3/\gamma_2$. In contrast, larger admixtures of perturbing channels in the $\frac{1}{2}[\frac{5}{2}]_2^o$ channel result in lifetime decreases of the $\frac{1}{2}[\frac{5}{2}]_2^o$ states at the two regions of ν_1. For clarity, the predicted lifetimes of the $6pnd\ \frac{1}{2}[\frac{5}{2}]_2^o$ $(n=14-33)$ levels around the second perturber are listed in Table III. The other theoretically predicted $6pnd\ \frac{1}{2}[\frac{5}{2}]_2^o$ and $\frac{1}{2}[\frac{3}{2}]_2^o$ states have the character of $(\nu_1)^3/\gamma_i$ and the lifetimes are easily evaluated with the parameters obtained here.

In summary, the MQDT parameters are obtained by fitting the Rydberg series $6pnd\ J=2$ energy levels of Pb I.

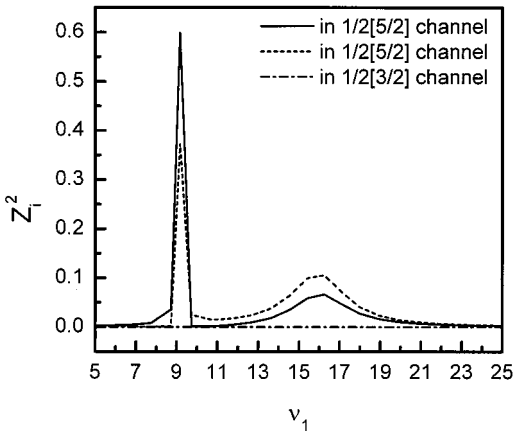

FIG. 2. Admixture coefficients $(Z_i^{(n)})^2$ of the $(3/2)[5/2]_2^o$ and $(3/2)[3/2]_2^o$ perturbing channels in the ν_1 region from 5 to 25.

These parameters are used to calculate channel admixture coefficients, from which the theoretical lifetimes of the levels are evaluated. It is obvious that the lifetimes of the $6pnd\ \frac{1}{2}[\frac{5}{2}]_2^o$ levels far from perturbers nearly follow the dependence $\tau\propto(\nu_1)^3$, while in the regions near perturbers the lifetime changes drastically because the wave-function admixtures of different channels are large in these regions. The agreement between the experimental and theoretical MQDT lifetimes is better than with those evaluated by the RHF method. We found that there are two perturbers in the Rydberg series studied. One is located at 58 517.71, and the other at 59 403.89 cm^{-1}. The lifetime perturbations near the second perturber are predicted by MQDT and await testing by experiment. To perform the extensive experimental measurements, lasers with shorter tunable wavelength and a higher-resolution detecting system are needed. We are now making efforts toward these measurements.

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