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Radiative lifetimes of Rydberg 6pnd J=2 states of Pb I by multichannel quantum defect theory*

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Energy levels of the odd-parity 6pnd J=2 Rydberg states of Pb I are analysed by the multichannel quantum defect theory (MQDT) in the frame of a five-channel three-limit calculation model. With optimal MQDT parameters, channel admixture coefficients are obtained and used to calculate the theoretical lifetimes of the levels by comparing to the previously measured lifetimes. The predicted lifetimes for higher-lying Rydberg states are given and discussed. These predicted lifetimes are very different from those obtained by the four-channel two-limit model previously used, which means that introduction of the additional interacting channel is important for studying the 6pnd J=2 Rydberg states of Pb I.

Keywords: radiative lifetime, Rydberg state, lead atom, multichannel quantum defect theory
PACC: 3270F, 3150, 3190

1. Introduction

The investigations on atomic Rydberg states remain an important research topic in atomic physics and laser spectroscopy.[1,2] Multichannel quantum defect theory (MQDT) has been extensively and successfully applied for the description of perturbed Rydberg series.[3−10] In our recent paper,[11] radiative lifetimes of the 6pnd J=2 Rydberg states of the Pb atom have been analysed and predicted by using the MQDT technique and all the experimental energy levels measured by Brown *et al*[12] and by Wood and Andrew.[13] The four-channel two-limit model for MQDT calculations, which was primarily employed by Brown *et al*[12] in their MQDT analysis with 60 energy levels of the 6pnd J=2 Rydberg states of Pb I, was used in Ref.[11] to fit the energy levels and then the wavefunctions of the levels were gained.

However, for the higher-lying energy levels from 59508.25 cm^{-1} to 59710.4 cm^{-1} measured in Ref.[12], the quantum defect values (modulo 1) of some levels have distinct changes from 0.225 to 0.295 with the increase of energy. This indicates that there exists another perturbing channel, which was not considered in Refs.[11, 12], to influence the 6pnd J=2 Rydberg states of Pb I. The perturber in this channel has a probability of influencing the lifetimes of the higher-lying levels around it. Therefore, a particular reinvestigation on this Rydberg series is necessary to be carried out in a five-channel MQDT model by introducing a new J=2 perturbing channel into the four-channel model used before. Detailed presentation of this new channel is given in Section 3. In this paper, we employ a five-channel three-limit model to perform MQDT analyses on energy levels of the 6pnd J=2 Rydberg states of Pb I. By using the MQDT wavefunctions obtained in this model, lifetimes of these states were calculated and compared with the previous results.

2. Basic theoretical methods

The theoretical methods and formulae for MQDT analyses and lifetime calculation of Rydberg levels are the same as those described in Ref.[11]. Detailed presentations of these methods can be found in the original papers.[3−5,14,15]

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3. Specification of the new perturbing channel

In the four-channel two-limit MQDT model, the 6pnd J=2 Rydberg states of Pb I are described with four interacting channels: the 6pnd $1/2[5/2]^o_2$ and 6pnd $1/2[3/2]^o_2$ channels converge to the first ionization limit 6p $^2P_{1/2}$ (59819.57 cm^{-1}) and the 6pnd $3/2[5/2]^o_2$ and 6pnd $3/2[3/2]^o_2$ channels to the second one 6p $^2P_{3/2}$ (73900.64 cm^{-1}).[11,12] The Lu–Fano plot calculated with optimal MQDT parameters obtained in Ref.[11] in this model is shown in Fig.1. The agreement between the theoretical fitted curves and experimental points is good except for some points in the region from ν_2=2.761 to 2.781, where ν_2 is the effective quantum number calculated with the second ionization limit. These points were not well fitted since the quantum defect values (modulo 1) of these levels are quite different in a large range from 0.225 to 0.295. This kind of quantum defect shift cannot be attributed to measurement errors. According to wavelength uncertainties of observed absorption lines of Pb I given by Brown *et al*,[12] quantum defect errors of these points are smaller than 0.012 except that the three levels 59625.1, 59640.5 and 59654.1 cm^{-1} determined from blended lines have larger errors. Therefore, we think that the quantum defect shift mentioned above is caused not by experimental errors but by the influence of another perturbing channel.

In experimental and theoretical researches of the odd-parity levels of Pb I, Dembczyński *et al*[16] found that the 6s6p^3 configuration strongly interacts with the 6pnd configurations, and especially that the state 6s6p^3 $^5S^o_2$ interacts with many 6pnd J=2 levels below the first ionization limit. In addition, the relativistic Hartree–Fock calculations by Pejcev *et al*[17] and the linked-parameter level fitting calculations by Dembczyński *et al*[16] indicated that the 6s6p^3 $^5S^o_2$ level is located below the first limit. Thus the 6s6p^3 $^5S^o_2$ state has a large probability to influence the energy positions and other properties of the 6pnd J=2 states around it.

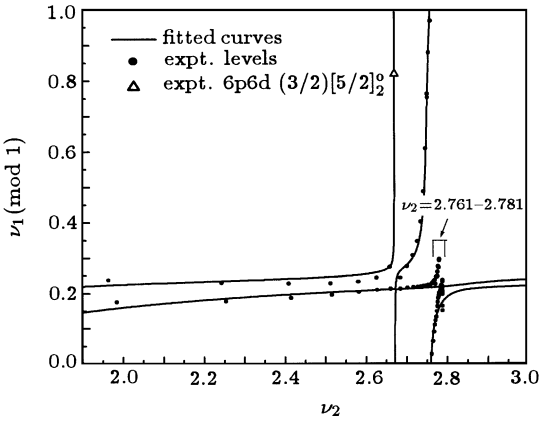


Fig.1. Lu–Fano plot obtained in the four-channel two-limit model for the 6pnd J=2 levels of Pb I.

Based on the above discussion, the additional perturbing channel 6s6p^2np $^5S^o_2$ should be taken into account in MQDT analyses of the 6pnd J=2 Rydberg series of Pb I. The ionization limit of the 6s6p^2np $^5S^o_2$ channel is taken as 73905.00 cm^{-1} which corresponds to the 6s6p^2 $^4P_{5/2}$ state of Pb II.[18]

4. Results and discussion

By introducing the additional 6s6p^2np $^5S^o_2$ channel, the MQDT model for analyses of the 6pnd J=2 Rydberg states of Pb I should be a five-channel three-limit model, in which five interacting channels are labelled with eigen-channels as

6pnd	6pnd	6pnd	6s6p^2np	6pnd
$(1/2)[5/2]^o_2$	$(1/2)[3/2]^o_2$	$(3/2)[5/2]^o_2$	$^5S^o_2$	$(3/2)[3/2]^o_2$

Ninety experimental energy levels of the 6pnd J=2 Rydberg states used in MQDT calculation are in the range from 45443.17 to 59787.87 cm^{-1} reported by Brown *et al*[12] and by Wood and Andrew.[13] All these levels are located below the first ionization limit. Here only the energy dependence of the eigen-quantum defect μ_α is considered with $\mu_\alpha = \mu^0_\alpha + \varepsilon\mu^1_\alpha$, where ε is the energy measured from the first ionization limit. The energy dependence of the parameter θ_{ij}, which is the rotation transformation angle between two interacting channels, is neglected, as is done in general. By fitting theoretical values of energy level to experimental ones, we obtained the optimal MQDT parameters as listed in Table 1. The Lu–Fano plot obtained in the five-channel three-limit model is shown in Fig.2. By comparison with Fig.1, it is clear that the MQDT fitting in the present model is much better than that in the four-channel two-limit model. When the lowest two levels 6p6d $(1/2)[5/2]^o_2$ and 6p6d $(1/2)[3/2]^o_2$ are excluded, the root mean square (rms) deviation between the theoretical and experimental energy levels in the present model is 2.87 cm^{-1}, while the rms deviation in the old model is 6.01 cm^{-1}.

Two vertical branches with larger ν_2 values in

Table 1. MQDT parameters for the odd-parity J=2 bound levels of Pb I.

Channel	1	2	3	4	5
$\|i\rangle$	$(1/2,\ 5/2)^{\rm o}_2$	$(1/2,\ 3/2)^{\rm o}_2$	$(3/2,\ 5/2)^{\rm o}_2$	$(5/2,\ 1/2)^{\rm o}_2$	$(3/2,\ 3/2)^{\rm o}_2$
I_i	59819.57	59819.57	73900.64	73905.00	73900.64
$\|\bar{\alpha}\rangle$	6pnd $(1/2)[5/2]^{\rm o}_2$	6pnd $(1/2)[3/2]^{\rm o}_2$	6pnd $(3/2)[5/2]^{\rm o}_2$	6s6p^2np $^5S^{\rm o}_2$	6pnd $(3/2)[3/2]^{\rm o}_2$
μ^0_α	0.2299	0.1709	0.3395	0.2995	0.2137
μ^1_α	0.2449	−0.0900	−0.6687	−1.2632	−0.1380
	0.7151	0.0582	−0.1386	0.2543	−0.6335
	0.0	0.8089	0.0695	−0.5598	−0.1656
$U_{i\alpha}$	0.1004	−0.0487	0.9879	0.0751	−0.0773
	−0.2413	0.5627	0.0	0.7850	0.0945
	0.6483	0.1527	0.0	0.0	0.7459

Fig.2 arise from the perturbation of the 6p6d $(3/2)[3/2]^{\rm o}_2$ and 6s6p^3 $^5S^{\rm o}_2$ levels, respectively. According to the analyses given by Dembczyński *et al*,[16] the 6s6p^3 $^5S^{\rm o}_2$ level is located at around 59400 cm^{-1}, which means that the 6s6p^3 $^5S^{\rm o}_2$ state influences the levels in the first one of the two vertical branches mentioned above.

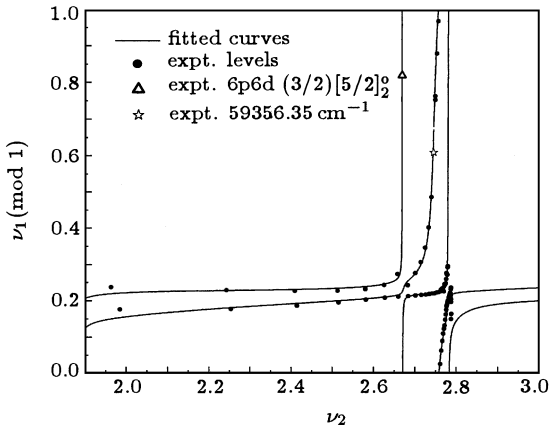


Fig.2. Lu–Fano plot obtained in the five-channel three-limit model for the 6pnd J=2 levels of Pb I.

The formula of radiative decay rate for the odd-parity 6pnd J=2 Rydberg states can be expressed as [15]

$$\Gamma_n = \left(Z_1^{(n)}\right)^2 \frac{\gamma_1}{\left(\nu_1^{(n)}\right)^3} + \left(Z_2^{(n)}\right)^2 \frac{\gamma_2}{\left(\nu_1^{(n)}\right)^3} + \left(Z_3^{(n)}\right)^2 \Gamma_3 + \left(Z_4^{(n)}\right)^2 \Gamma_4 + \left(Z_5^{(n)}\right)^2 \Gamma_5,$$

where γ_1, γ_2, Γ_3, Γ_4 and Γ_5 are radiative rate parameters in relation to the 6pnd $(1/2)[5/2]^{\rm o}_2$, $(1/2)[3/2]^{\rm o}_2$, 6pnd $(3/2)[5/2]^{\rm o}_2$, 6s6p^2np $^5S^{\rm o}_2$ and 6pnd $(3/2)[3/2]^{\rm o}_2$ channels, respectively; ν_1 is the effective quantum number in the first limit. By fitting the theoretical lifetimes to 16 experimental lifetimes we measured previously [19] with MQDT admixture coefficients, we obtain the radiative rate parameters as follows: γ_1=7.789×10^8 s^{-1}, γ_2=31.596×10^8 s^{-1}, Γ_3=2.948×10^8 s^{-1} and Γ_4=1.387×10^8 s^{-1} and Γ_5=29.699×10^8 s^{-1}. Using these parameters, lifetime calculations of the 6pnd J=2 states are extended to the level 59787.87 cm^{-1}, which is the highest experimental level measured in Ref.[12] and 116 lifetimes are obtained. The calculated lifetimes and the comparison with previously measured lifetimes are shown in Fig.3. As seen from Fig.3, the good agreement between the theoretical and experimental lifetimes indicates that MQDT parameters and admixture coefficients obtained in the five-channel three-limit model are reliable for describing the 6pnd J=2 Rydberg states. Due to the introduction of the new interacting channel and corresponding MQDT parameters, the rms error 9.42 ns between the theoretical and experimental lifetimes is smaller than the rms error 17.29 ns in the old model.[11]

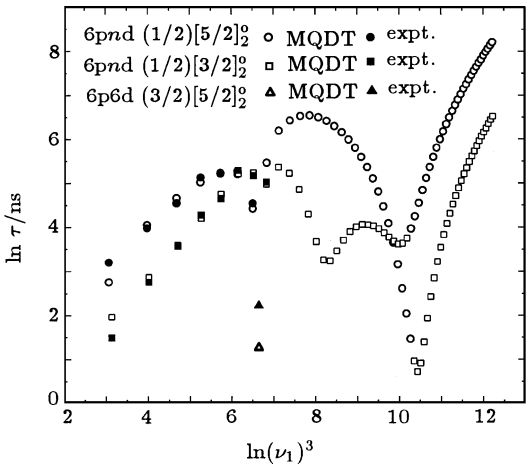


Fig.3. MQDT theoretical lifetimes and comparison with experimental values for the 6pnd J=2 states of Pb I.

For the predicted lifetimes, there exist certain prominent differences when using the two different models, as can be seen from the comparison between Fig.3 in this paper and the results in Ref.[11]. In the five-channel model, the lifetimes of levels in both 6pnd

$(1/2)[5/2]^o_2$ and 6pnd $(1/2)[3/2]^o_2$ perturbed channels in the region of $\ln(\nu_1)^3 > 8$ decrease greatly due to the interaction of the perturbing channel newly introduced here. It should be noted that the designations of levels shown in Fig.3 are determined from our MQDT admixture coefficients. For the levels in the 6pnd $(1/2)[5/2]^o_2$ and $(1/2)[3/2]^o_2$ channels, the admixture coefficients of other channels calculated in the present model are shown in Figs.4 and 5, respectively. The two figures indicate that the 6p6d $(3/2)[3/2]^o_2$ perturbing channel has been mixed into the 6pnd $(1/2)[3/2]^o_2$ channel more than into the 6pnd $(1/2)[5/2]^o_2$ channel. In contrast, the $6s6p^2np\ ^5S^o_2$ channel only has influence on levels in the 6pnd $(1/2)[3/2]^o_2$ channel. In our calculations, the maximum admixture of the $6s6p^3\ ^5S^o_2$ state is 15.2% to the level 59356.35 cm^{-1} (ν_1=15.39, ν_2=2.747, eigenvector purity=81.0%), which is in good agreement with the semiempirical result 14.8% to the same level gained by Dembcyński *et al.*[16]

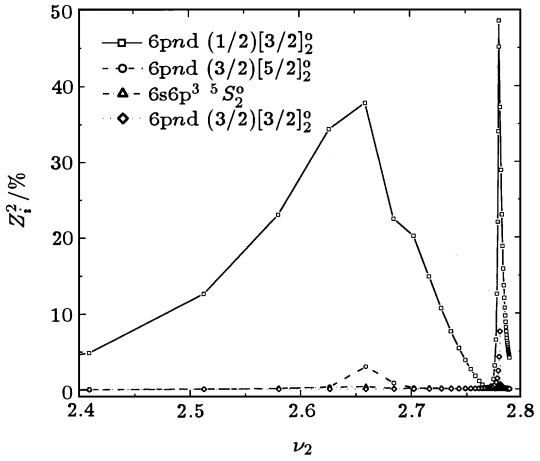


Fig.4. Admixture coefficients of the other channels to the 6pnd $(1/2)[5/2]^o_2$ channel.

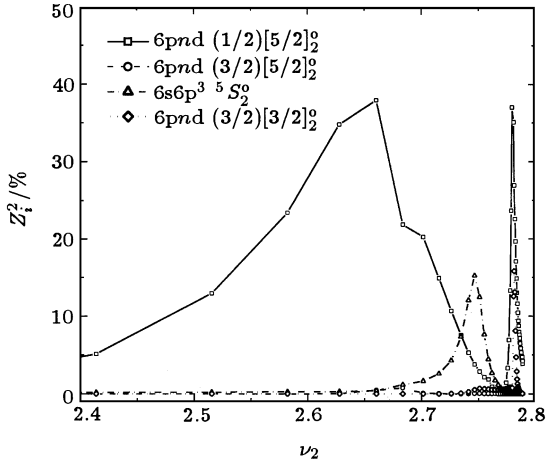


Fig.5. Admixture coefficients of the other channels to the 6pnd $(1/2)[3/2]^o_2$ channel.

As can be seen in Fig.3, sharp changes in the respective lifetimes of the two perturbed channels occur near 59703 cm^{-1} (ν_1=30.7 or $\ln(\nu_1)^3$=10.27). The reason for this is that, in the region of ν_1=27.7–30.7 ($\ln(\nu_1)^3$=9.96–10.27), the admixture coefficients of the 6p6d $(3/2)[3/2]^o_2$ perturbing channel to the 6pnd $(1/2)[5/2]^o_2$ channel are larger than those to the 6pnd $(1/2)[3/2]^o_2$ channel. Therefore, due to more admixture of the short-lifetime characteristic of the 6p6d $(3/2)[3/2]^o_2$ channel, the lifetimes of 6pnd $(1/2)[5/2]^o_2$ levels in this region are smaller than those of the 6pnd $(1/2)[3/2]^o_2$ levels.

Figure 3 shows that, except for a few states in the region of $\ln(\nu_1)^3 < 6$, the lifetime values of the 6pnd J=2 Rydberg states in the energy region studied do not have the pure Rydberg state character of $(\nu_1)^3/\gamma_i$ (i=1, 2) owing to the perturbation of the new channel introduced in this paper. The theoretically predicted lifetimes obtained here await testing by experiment.

It should be pointed out that the designations of levels above 58666.94 cm^{-1} determined from the admixture coefficients in the present five-channel model are entirely opposite to those determined in the four-channel model. This results from the introduction of the $6s6p^2np\ ^5S^o_2$ interacting channel. Clearly, the choice of MQDT channel model is important for the theoretical designations of level notations for the levels having no labels.

5. Conclusions

In conclusion, we have introduced an additional perturbing channel $6s6p^2np\ ^5S^o_2$ in MQDT investigation on energy levels and lifetimes of the odd-parity 6pnd J=2 Rydberg states of Pb I. The MQDT parameters are obtained by fitting the theoretical energy levels to experimental ones. Using channel admixture coefficients calculated from these parameters, we have obtained the theoretical lifetimes of the levels by fitting to the measured lifetimes. The agreement between the theoretical and experimental results of energy levels and lifetimes obtained in the five-channel three-limit model is better than that in the four-channel two-limit model. It is found that due to the influence of the perturbing channels the lifetimes of the higher-lying 6pnd J=2 Rydberg states below the first ionization limit are not directly proportional to $(\nu_1)^3$, as is the character of the pure Rydberg state. Such a distinct influence on lifetimes coming from the newly introduced channel is unexpected. Therefore,

to introduce this additional interacting channel and study its influence on energy levels, lifetimes and other properties of the 6pnd J=2 Rydberg states of Pb I are important for understanding the characters of lead atom. It would be also interesting to check the lifetime values of higher-lying 6pnd J=2 Rydberg states of Pb I by experiment and compare them with the theoretical results in this paper.

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