Rydberg states and spin-orbit coupling of the thallium atom

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Using the resonance-enhanced multiphoton-ionization technique, we have identified the $n^2P_{1/2}(7 \leq n \leq 20)$, $n^2P_{3/2}(7 \leq n \leq 23)$, and $n^2F_{3/2,3/2}(5 \leq n \leq 21)$ Rydberg states following photodissociation of TII. The $n^2P_{1/2,3/2}$ spin-orbit splitting can be resolved and well compared to its theoretical counterpart with an accuracy of $\pm 3.5\%$. In terms of photoionization-cross-section prediction, we have also satisfactorily interpreted the discrepancy for the relative ionization intensity, via the respective intermediate $n^2P_{3/2}$ and $n^2P_{1/2}(n \leq 11)$ doublets, from their statistical weight.

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I. INTRODUCTION

The resonance-enhanced multiphoton-ionization (REMPI) technique has been widely employed to probe the high-lying states of an atom or a molecule, and thereby numerous new states are identified [1,2]. These Rydberg states are difficult to probe with conventional optical spectroscopy. For detecting a high-lying atom, for instance, the fluorescence method will become less sensitive with increase of principal quantum number $n$, since the transition oscillator strength varies in proportion to $n^{-3}$ [1], while REMPI can feasibly ionize the excited atoms and monitor the resulting ions with a detection efficiency close to unity. Therefore, in this paper we attempt to employ this technique to characterize the Rydberg states of TI atoms, which will result from the photodissociation of TII. Mirza and Duley have also conducted a similar REMPI experiment with pure TI sample, and identified the TI $nP$ and $nF$ Rydberg states up to $n=21$ and 20, respectively [3]. However detailed work on the resolution of the $n^2P_{1/2}$ and $n^2P_{3/2}$ spin-orbit splitting and the characterization of their relative intensity in the ionization process has not yet occurred. This motivates us to carry out this experiment.

In our work, a Nd:YAG (where YAG denotes yttrium aluminum garnet) laser-pumped dye laser has been employed to produce TI ($6^2P_{1/2}$) and TI* ($6^2P_{3/2}$) fragments following a one- or two-photon dissociation of TII, and subsequently to ionize these atoms through their intermediate $nP$ or $nF$ Rydberg states by a (2 + 1)-REMPI process. In this manner, we have identified the $n^2P_{1/2}(7 \leq n \leq 20)$, $n^2P_{3/2}(7 \leq n \leq 23)$, and $n^2F_{3/2,3/2}(5 \leq n \leq 21)$ Rydberg series, and have resolved the spin-orbit couplings for the $nP$ fine-structure states; the latter splittings are satisfactorily compared to their theoretical counterparts based on a one-valence-electron, single-configuration model. Finally, in terms of the ionization-cross-section prediction, we have interpreted the relative intensity for the ionization process via the intermediate $n^2P_{1/2}$ and $n^2P_{3/2}$ doublets.

II. EXPERIMENT

The reagent TII, from purchase without further purification, was deposited in the finger reservoir of a six-armed quartz cell, which was enclosed in a firebrick oven. Through the top arm, the cell was connected to a vacuum line and evacuated below $10^{-5}$ Torr before operation. The temperature of the cell body was maintained around 593–633 K with an accuracy of $\pm 2$ K, about 80 K higher than that of the sample reservoir to prevent the TII vapor from condensing onto the windows. The resultant TII vapor pressure corresponded to $10^{-4}$–$10^{-3}$ Torr (or $3 \times 10^{12}$–$2 \times 10^{11}$ cm$^{-3}$) [4], and the number density of the fragmented TI was estimated to be $10^{10}$–$10^{11}$ cm$^{-3}$ [5]. A similar oven design as in this work has been reported in detail elsewhere [6,7].

As a radiation source for conducting both photodissociation and ionization processes, a Nd:YAG laser-pumped dye laser, having a bandwidth of 0.025 nm at 400 nm, a pulse duration of 5–8 nsec, and a repetition rate of 10 Hz was tuned ranging from 600 to 280 nm. The uv-wavelength portion was accessible through the operation of a frequency-doubled potassium dihydrogen phosphate (KDP) crystal device. The laser energy varied from 1–30 mJ throughout the experiment. Following photodissociation of the TII, the focused, impinging laser beam can then successively ionize the fragmented TI atoms through a selective intermediate $nP$ or $nF$ Rydberg state. The multiphoton-ionization process was considered to dominate over the other ionization processes in our case with a TII vapor pressure as low as $10^{-3}$–$10^{-4}$ Torr [8]. However, when $n$ increases, the energy defect between the Rydberg states ($n > 14$) and the ionization threshold becomes smaller than the thermal energy $kT$. Thus the contribution of the thermal ionization may then become significant.

The produced TI$^+$ ions were collected with a pair of voltage-biased nichrome probes, which were set about 1 cm apart. The ion current was amplified with a current amplifier prior to input to a boxcar integrator for signal
processing. The resulting ion signal was displayed on an
oscilloscope or stored in a microcomputer for further
analysis.

III. RESULTS AND DISCUSSION

A. Rydberg state identification

The Rydberg states of Tl atoms, as fragmented to the
ground or spin-orbit excited states following photodissoc-
iation of TlI, were probed using a (2 + 1)-REMPI tech-
nique. A portion of the ionization spectra is shown in
Fig. 1. The $n^2P_{1/2}$($7 \leq n \leq 20$), $n^2P_{3/2}$($7 \leq n \leq 23$), and
$n^2F(5 \leq n \leq 21)$ Rydberg series are identified; the results,
which are compared well to those reported elsewhere
[3,9], are listed in Table I.

The term energies obtained can be essentially charac-
terized using the Rydberg formula [1,2]. By taking the
derivative of the formula with respect to the principal
quantum number $n$, one obtains

$$\frac{dE_n}{dn} = \frac{2\hbar \delta}{(n-\delta)^2},$$  \hspace{1cm} (1)

where $\hbar \delta$ is the Rydberg constant, $\delta$ is the quantum
defect, and $n-\delta$ denotes the effective principal quantum
number which can be related to the term energy $E_n$ by
the expression

$$n-\delta = n^* - \frac{Z_a}{E_n^{1/2}}.$$  \hspace{1cm} (2)

For a neutral atom, the charge of the atomic core $Z_a$
is equal to unity. As $n$ becomes large, the left-hand side
of Eq. (1) can be replaced by $\Delta E_n / \Delta n$ approxi-
mately; $\Delta E_n$ is the term energy difference between two
successive states. The resulting log-scale plot of $\Delta E_n / \Delta n$
for the $n^2P_{1/2}$, $n^2P_{3/2}$, and $n^2F$ Rydberg series against $n^*3$ is
shown in Fig. 2; the slope is measured to be $-1$, indicat-
ing that the Rydberg state assignment should be reliable.
Especially for large $n$, the term energy can be well
characterized by Eq. (1). The values of $n^*$ and $\delta$ as func-
tions of $n$ are also listed in Table II. As expected, the $n$
dependence of $\delta$ is found to be invariant.

We can estimate the photon numbers involved in the
photodissociation process by the simple formula
$n\hbar \nu \geq \Sigma E_i + E_d$. $E_i$ indicates the term energy
of the fragmented atoms and $E_d$, the dissociation energy
of TlI. Since the threshold wavelength of photodissocia-
tion into the channel of TlI*($6^2P_{1/2}$) and I($5^2P_{3/2}$) is $\sim 339$ nm
[10], the process of producing a TlI*($6^2P_{1/2}$) fragment
upon irradiation at wavelengths $> 500$ nm requires two-
photon absorption. In this wavelength domain ($500$–$600$) nm, we have identified the $nP$, $8 \leq n \leq 12$ and
$nF$, $5 \leq n \leq 9$ structures with TlI*($6^2P_{3/2}$) as the lower
state of the ionization transition. The assignment is con-
sonst with those results with the ground TlI($6^2P_{1/2}$)
fragment as the initial state in the related transition.

B. Spin-orbit coupling in the $nP$ doublets

The results of spin-orbit splitting observed in the $nP$
doublets are listed in Table III. Spin-orbit coupling
mainly results from the interaction between the magnetic
field of the circulating electron and the magnetic moment
associated with the electron spin. For simplicity, we
adopt a model suitable for a one-valence-electron, single-
configuration condition. The related equation is given ex-
plicitly in the following [11,12]:

$$\Delta E_{1/2,3/2} = \langle \Psi_p | a(r) | \Psi_p \rangle$$

$$= \alpha^2 \left( \frac{dn^*}{dn} \right) \frac{Z_i^2}{n^*3} \left( \frac{Z_i^2 H_v R}{l(l + 1)(l + \frac{1}{2})} \right).$$  \hspace{1cm} (3)

Here, $\alpha$ denotes the fine-structure constant, $Z_i$ the
effective nuclear charge, and $H_v$ a relativistic correction.
The value of $H_v$ is almost unity for a light atom, but
varies as an exponential-like function for the heavy atoms
with atomic number $Z > 50$. Given an appropriate value
of 1.2 for $H_v$ adopted from Ref. [13], the best fit of simu-
lation with Eq. (3) to the splitting observation of the $nP$
doublets leads to a value of 75.4 for $Z_i$. We believe that
our result is more precise than a previous value of 75,
determined by taking into account only the lowest excited
state [14]. The provision of $Z_i$ is important in deter-
mining the hyperfine structure; its value for the Tl atom
is on the order of $10^{-1}$ cm$^{-1}$, which cannot be resolved
in this work.

To our surprise, as one can see in Table III, the simple
model can be suitable for interpreting the spin-orbit split-
ting observed in the Tl $nP$ doublets with an accuracy of
$\pm 3.5\%$ except for $n = 20$. The theoretical prediction
in Table III is very consistent with our results, but slightly
deviates from the data reported previously [3,9]. This
fact may also confirm the reliability of our spectral as-
ignment of the Rydberg states. Nevertheless, the model
must be refined apparently to deal with the more compli-
cated observation. A dedicated model, for instance, tak-
ing into account the exchange polarizability of the core
electrons with the valence electron, has been employed
successfully for interpreting the negative spin-orbit con-
stant found in the Rb atom [15].
TABLE I. Term energy for \( nP_{1/2, 3/2} \) and \( nF_{5/2, 7/2} \) Rydberg states of the Tl atom in units of \( \text{cm}^{-1} \). The uncertainty is within \( \pm 0.4 \text{ cm}^{-1} \).

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It is interesting to note that the intensity ratio of ionization transition via the respective intermediate \( n^2P_{3/2} \) and \( n^2P_{1/2} \) doublets behaves abnormally from the ratio of their statistical weights. As shown in Fig. 3, the \( n^2P_{3/2} \) to \( n^2P_{1/2} \) ratio of ionization intensity at \( n \leq 14 \) is smaller than 2, but becomes larger with increasing \( n \). A similar result has been found in Rb, giving a ratio of 5.9 at large \( n \) [16]. The intensity ratio of Hg \( ^+D_{5/2} \) to \( D_{3/2} \) ionic states was also found to be 2.0 at 548 Å and 1.37 at 304 Å, showing discrepancy from 1.5 [17,18]. Such abnormal phenomena have been theoretically interpreted, for instance, by taking into account configuration interaction [19] or core polarizability exchange [20]. The intensity of ionization transition can be assumed to be a product of the transition probabilities between the first-step transition from the Tl \( 6^2P_{1/2} \) (or \( 6^2P_{3/2} \)) to \( n^2P_{3/2} \) or \( n^2P_{1/2} \) intermediate state as well as the second-step transition from the \( n^2P_{3/2} \) or \( n^2P_{1/2} \) to continuum state. Since the relative intensity for the first-step transition depends upon the statistical weights of the \( nP \) fine-structure components, therefore, the abnormal intensity ratio observed above is subject to the dynamic variation of the matrix element related to the second-step ionization transition. The ionization transition probability, which is proportional to the photoionization cross section, can be expressed as \( |\langle \Psi_p | r | \Psi_p \rangle|^2 \). The initial electronic wave function \( \Psi_p \) denotes the Tl \( n^2P_{3/2} \) or \( n^2P_{1/2} \) state; the final continuum wave function \( \Psi_c \) denotes the

FIG. 2. Logarithmic plot of the \( n^* \) dependence of \( \Delta E/\Delta n \). \( \Delta E \) indicates the term energy difference (in \( \text{cm}^{-1} \)) between two successive states. The symbol ■ indicates \( n^2P_{1/2} \), + indicates \( n^2P_{3/2} \), and ◊ indicates \( n^2F_{5/2, 7/2} \) Rydberg states.

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TABLE III. Spin-orbit splitting in units of cm$^{-1}$ for the $n^2P_{1/2,3/2}$ doublets. The experimental uncertainty is within $\pm 0.6$ cm$^{-1}$.

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cross section. The wave function of the valence electron for the $n^2P_{3/2}$ spin-orbit excited state is expected to be repelled slightly further from the nucleus with respect to the $n^2P_{1/2}$ state, and accordingly experiences the overlap with the continuum wave function of the ionic state prior to that of the $n^2P_{1/2}$ state. If the photoionization cross section associated with the $nP$ state rises with the excess energy, that suggests the ionization transition intensity via the $n^2P_{3/2}$ intermediate state should experience enhancement prior to that via the $n^2P_{1/2}$ state, and accordingly the $n^2P_{3/2}$ to $n^2P_{1/2}$ ratio of ionization intensity becomes larger than their statistical-weights ratio. Analogously, a decrease for the photoionization-cross-section prediction as a function of the excess energy may explain the behavior of the intensity ratio smaller than 2. Such an interpretation has been successfully applied to the ionization-intensity ratio related to the subshell electron configuration, which can be probed by photoelectron spectroscopy [17,18].

In the following, we calculate the photoionization cross section of $nP$ as a function of the excess energy using the quantum-defect method, which was first developed by Burgess and Seaton [22] and later modified by Peach [23]. The photoionization cross section can be expressed explicitly as follows [23]:

$$a_\alpha = \frac{4\pi a_0 e^2}{3} \left| \frac{Z_s^2}{u} + Z_e e' \right|$$

$$\times \sum_{r'=1+1} C_{r'} \left[ \int_0^{\infty} P_{si}(r) r G_{k,l'}(r) dr \right]^2,$$

where $\alpha$ is the fine-structure constant and $a_0$ is the Bohr radius. $P_{si}(r)$ indicates the initial radial wave function of the bound state, and $G_{k,l'}(r)$, the final radial wave function of the continuum state. $e'$ denotes the energy (in units of rydbergs) of the ejected electron, which has angular momentum $l'$, $k$ is defined to be $(Z^2_e e')^{1/2}$, and $Z_s$ is the charge of atomic core and equal to unity for neutral atoms. $v(t=\infty)$ is the effective quantum number and is defined in Eq. (2). The coefficients $C_{r'}$ are related to the angular momentum and spin quantum number of the ini-

![FIG. 3. The n dependence of the ionization intensity ratio via the intermediate n^2P_1/2 and n^2P_3/2 doublets. The symbol ○ indicates that Tl (6^2P_1/2) is used as the lower state in the ionization transition, and × indicates that Tl (6^2P_3/2) is the lower state of the related transition.](image)

![FIG. 4. Calculation of the photoionization cross section of the Tl n^2P states (n=8,9,11) as a function of excess energy (in units of rydbergs) over the ionization threshold.](image)
tial and final states. For calculation of these coefficients the reader is referred to Ref. [22]. According to Eq. (4), we follow the method developed by Peach [23] to calculate the photoionization cross section as a function of the excess energy. The result is shown in Fig. 4, revealing that the cross section decreases with increasing the excess energy, as $n \leq 11$. The fact is consistent with the interpretation described previously for the ratio smaller than 2. For large $n$ ($n \geq 14$), however, we cannot satisfactorily apply this model for several reasons. First of all, the accuracy inherent in this model becomes very poor as the excess energy (or $n$) becomes large [22,23]. Second, the fine-structure splittings become so small about the order of 10 cm$^{-1}$ for large $n$ that the radial wave-function overlap between the respective fine-structure doublets and the ionization continuum hardly seems to resolve spatially. Thus interpretation with the concept described above becomes insignificant. Finally, as mentioned in the preceding section, the photoionization process no longer prevails for large $n$; the ionization mechanism may be partly contributed from the thermal ionization channel. Accordingly, it will be less significant with the photoionization cross-section prediction to account for the ionization ratio at large $n$.

In summary, using the REMPI technique, we have achieved identification of the $n^2 P_{1/2} (7 \leq n \leq 20)$, $n^2 P_{3/2} (7 \leq n \leq 23)$, and $n^2 F_{5/2,7/2} (5 \leq n \leq 21)$ Rydberg states. We have also carried out a model calculation, which is consistent with the observation of spin-orbit splitting with an accuracy $\pm 3.5\%$. For small $n$, the $n^2 P_{3/2}$ to $n^2 P_{1/2}$ ratio of ionization intensity can be satisfactorily interpreted in terms of photoionization-cross-section prediction.

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