Stark Spectroscopy for Studying the Parity Nonconservation in Atomic Samarium

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Abstract

Spectroscopic study of atomic samarium (Sm) has been performed as a preliminary investigation for measuring the parity nonconservation (PNC) in atoms. In the first stage of the experiments, we have systematically measured seven $E_1$ transitions from the $4f^66s^2^7F_{0-6}$ to $4f^66s6p^7G_{1-6}$ levels by atomic-beam laser spectroscopy. The 15650.55-cm$^{-1}$ ($4f^66s6p^7G_1$) level, which has a close-lying opposite-parity neighbor, is one of candidates for PNC measurements in Sm. We have observed the hyperfine splittings and isotope shifts without external field. The hyperfine coupling constants $A$ and $B$ of $4f^66s6p^7G_{2-6}$ levels have been newly determined for $^{147}$Sm and $^{149}$Sm. King plot analysis for the isotope shift has led to the specific mass shift and field shift. Stark splittings of the transitions have been measured under external electric field. The tensor polarizabilities $\alpha_2$ have been precisely determined for the $4f^66s6p^7G_{1-3}$ levels. As a result, the isotope dependence of $\alpha_2$ has been clearly observed for the 15650.55-cm$^{-1}$ for the first time. This phenomenon is possibly useful to refine the calculation of the wave function for the PNC measurements. Zeeman splittings of the transitions have been measured under external magnetic field. The gyromagnetic ratios $g$-values have been precisely determined for the $4f^66s6p^7G_{2-6}$ levels.

As a next stage of the experiments, we have tried to observe the Stark-induced $E1$ transitions with optical double-resonance technique. We have performed electric discharge experiments to search suitable high-lying odd-parity levels. While the pump laser was locked to an $E1$ transition from the $0$-cm$^{-1}$ ($4f^66s^2^7F_0$) to 15650.55-cm$^{-1}$ levels, the probe laser was tuned to the parity forbidden transitions under electric field. In the probe transition from the 15650.55-cm$^{-1}$ to 28233.08-cm$^{-1}$ levels ($4f^55d6s^2 J = 1$), we have successfully observed the Stark-induced $E1$ transition.
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Chapter 1

Introduction

The phenomenon of parity nonconservation (PNC) in atoms, which is sometimes called as atomic parity violation (APV), is induced by the weak interaction between the atomic electrons and nucleons through $Z^0$ boson [1, 2]. It gives rise to the mixing between the levels of opposite parity. This unique phenomenon has attracted many physicists from the aspect of particle physics as well as atomic physics. The weak charge $Q_W$, which is an observable of the PNC measurement in atoms, is sensitive to new physics beyond the standard model [3]. It has become an important tool in exploring physics beyond the standard electroweak model.

Bouchiat and Bouchiat pointed out that the PNC effects in atoms are enhanced in heavy elements, that is the dependence of atomic number – roughly proportional to $Z^3$ [1]. Thereafter, several precise PNC measurements have been reported for the heavy elements such as cesium ($^{58}$Cs) [4, 5], thallium ($^{81}$Tl) [6, 7], lead ($^{82}$Pb) [8] and bismuth ($^{83}$Bi) [9–11]. The most accurate value of $Q_W$ is given in Cs experiment at the present time [5, 12]. Rosner has recently reviewed the precision electroweak tests [13] including several reanalysis of Cs. The precisions in other elements except for Cs are not sufficient yet to provide a strong constraint on the new physics. The uncertainty from the atomic theory is one of the causes for large ambiguity of $Q_W$. It is now important to confirm the Cs results using other elements and to increase the accuracy.

Dzuba et al. have proposed that the PNC measurements using the rare-earth atoms could increase the accuracy [14]. Since the level structure of rare-earth atoms is complicated, they have nearly degenerate levels of opposite parity. A large PNC enhancement is expected in such close-lying levels. In addition, since they have many stable isotopes, it is expected that the uncertainty from the atomic theory is eliminated using the experimental results from isotope chain [14].

Among the rare-earth atoms, samarium ($^{62}$Sm) has interested the physicists at the early stage of the PNC measurements [14, 15]. One of the candidates for the PNC measurements is a pair of nearly degenerate levels where the energy separation is only 10.75 cm$^{-1}$. In contrast, the energy separation between the parity mixing levels is about 7500 cm$^{-1}$ in case of Cs. Moreover, Sm has seven naturally occurring isotopes while Cs has only one. Several measurements have been performed with optical rotation method to investigate possible large PNC enhancement [16, 17]. According to their conclusions, the smallness of $E1$ amplitude seems to have prevented an observation of possible enhancement of PNC effects if any. In order to resolve the problem in Sm, we propose new PNC measurement, which is based on Stark interference technique, by means of the optical double-resonance technique.
CHAPTER 1. INTRODUCTION

The purpose of the present work is to obtain precise spectroscopic data of the 15650.55-cm$^{-1}$ ($4f^66s6p^7G_1$) level, which has a close-lying neighbor, and to investigate the feasibility of PNC measurement with optical double-resonance technique. In Chapter 2, general theoretical descriptions of PNC in atoms are given. In Chapter 3, we describe our plan of PNC measurement in Sm taking into account the previous PNC measurements performed by other groups. The overview of our experiments is given in Chapter 4. It also includes experimental apparatus and laser-frequency locking-system. In Chapter 5, we describe the spectroscopic study of $4f^66s6p^7G$ term as the first stage of the experiments. The description of electric discharge experiments is given in Chapter 6. The observation of Stark-induced $E1$ transition is described in Chapter 7. In Chapter 8, we examine the obtained results and discuss the feasibility of the PNC measurement in Sm with optical double-resonance technique.
Chapter 2

Theoretical Descriptions

2.1 Atomic Parity Non-conservation

2.1.1 Neutral Weak Current Interaction in Atoms

In an atomic system, the PNC effect arises primarily from the interference between the neutral weak and electromagnetic interactions that couple a valence electron to the nucleus (Fig. 2.1). An orbital electron interacts with a nucleon by exchanging a massive $Z^0$ boson. The electronic and nucleonic weak neutral currents consist of vector and axial vector components [2,18]:

$$J_e = V_e + A_e, \quad J_N = V_N + A_N.$$  \hspace{1cm} (2.1)

The interaction Hamiltonian contains both scalar and pseudoscalar parts:

$$H_{\text{weak}} \simeq (V_e + A_e) \cdot (V_N + A_N)$$

$$= (V_eV_N + A_eA_N) + (A_eV_N + V_eA_N)$$

$$= H_S + H_P.$$  \hspace{1cm} (2.2)

The contribution of the parity conserving scalar term $H_S$ is negligible in the PNC experiment. Neglecting the momentum transfer, parity nonconserving pseudoscalar term $H_P$ can be written as

$$H_P = H_1 + H_2,$$  \hspace{1cm} (2.2)

![Figure 2.1: Electron-nucleus interactions in atom; (a) electromagnetic interaction with photon exchange and (b) weak interaction with $Z^0$ boson exchange.](image)
where

\[
H_1 = \sum_N A_e V_N = \frac{G_F}{\sqrt{2}} \sum_N \left( \psi_e^\dagger \gamma^\mu \gamma^5 \psi_e \right) C_{1N} \left( \bar{\psi}_N \gamma^\mu \psi_N \right),
\]

\[
H_2 = \sum_N V_e A_N = \frac{G_F}{\sqrt{2}} \sum_N \left( \psi_e^\dagger \gamma^\mu \psi_e \right) C_{2N} \left( \bar{\psi}_N \gamma^\mu \gamma^5 \psi_N \right).
\]

In these formula, \( G_F \) is the Fermi’s constants, \( \psi_e \) and \( \psi_N \) are the electron and nucleon Dirac spinors, respectively. The sums \( N \) are taken over all protons \( (p) \) and neutrons \( (n) \) in the nucleus. According to the standard model, the coupling coefficients are given by

\[
C_{1p} = \frac{1}{2} \left( 1 - 4 \sin^2 \theta_W \right) \sim 0.04,
\]

\[
C_{2p} = \frac{1}{2} g_A \left( 1 - 4 \sin^2 \theta_W \right) \sim 0.05,
\]

\[
C_{1n} = -\frac{1}{2},
\]

\[
C_{2n} = -\frac{1}{2} g_A \left( 1 - 4 \sin^2 \theta_W \right) \sim -0.05,
\]

where \( g_A \sim 1.25 \) is the axial vector coupling constant of neutron beta decay and \( \theta_W \) is the Weinberg mixing angle where \( \sin^2 \theta_W \sim 0.23 \). By taking the non-relativistic reduction of the nucleonic currents, we have

\[
H_1 = \frac{G_F}{\sqrt{2}} Q_W \left( \psi_e^\dagger \gamma^5 \psi_e \right) \rho(r),
\]

\[
H_2 = \frac{G_F}{\sqrt{2}} \sum_N C_{2N} \left( \bar{\psi}_e \gamma^5 \psi_e \right) \cdot \sigma_N \rho(r),
\]

where \( \rho(r) \) is a nucleon density distribution in the nucleus, and the \( \sigma_N \) is a nucleon spin operator. The weak charge \( Q_W \) is given by

\[
Q_W = \rho \left\{ Z (1 - 4 \sin^2 \theta_W) - N \right\}
\]

where \( Z \) and \( N \) are atomic and neutron numbers, respectively. Assuming

\[
\rho = M_W^2 / (M_Z^2 \cos^2 \theta_W) \simeq 1,
\]

with \( W \) and \( Z^0 \) boson masses, we have

\[
Q_W \simeq 0.08 Z - N.
\]

The value of \( Q_W \) is roughly equal to \( -N \). Since \( H_2 \) contains no enhancement factor \( Q_W \), the effect of \( H_2 \) is smaller than that of \( H_1 \) for heavy atom. Taking the non-relativistic limit, the non-relativistic potential for a single electron interacting with the nucleus is written as

\[
V_{\text{PNC}} = \frac{G_F}{4 \sqrt{2} m_e} Q_W \left\{ p_e \cdot \sigma_e, \rho(r) \right\},
\]

where \( m_e, p_e \) and \( \sigma_e \) are the mass, momentum and spin of the electron, respectively. The mixing matrix elements of \( V_{\text{PNC}} \) are nonzero only for atomic orbitals of opposite parity with nonvanishing value or gradient at the origin \( (s_{1/2}, p_{1/2} \text{ orbitals}) \). The mixing matrix element \( \langle s_{1/2} | V_{\text{PNC}} | p_{1/2} \rangle \) is roughly proportional to \( Z^2 Q_W \) [1].
2.1. ATOMIC PARITY NON-CONSERVATION

2.1.2 Stark Interference Method

We can classify the PNC measurements into two types. One is an optical-rotation method where the asymmetry of rotation of polarization plane is detected after the propagation of light through a matter. Another is a Stark-interference method. Since our measurement is based upon the Stark-interference method, we describe it only.

In order to simplify the situation, we consider the level scheme including three levels as shown in Fig. 2.2. It consists of a low-lying level \( |i^+\rangle \) and two high-lying levels \( |f^+\rangle, |n^-\rangle \), where the superscripts denote parity. The opposite-parity levels \( |f^+\rangle \) and \( |n^-\rangle \) are nearly degenerate. Through the electromagnetic interaction, an electric dipole (\( E_1 \)) transition is allowed between the opposite-parity levels \( |i^+\rangle \) and \( |n^-\rangle \). Between the same-parity levels \( |i^+\rangle \) and \( |f^+\rangle \), the \( E_1 \) transition is forbidden, but an magnetic dipole (\( M_1 \)) transition is allowed. Here we ignore the higher-order radiative transitions, such as an electric quadrapole (\( E_2 \)) transition. The respective transition amplitudes are given as

\[
A_{E1} = \langle n^- | d_z | i^+ \rangle, \\
A_{M1} = \langle f^+ | \mu_z | i^+ \rangle,
\]

where \( d_z \) and \( \mu_z \) are the electric dipole and magnetic dipole operators along the quantization axis, respectively. The amplitude ratio of \( A_{M1} \) to \( A_{E1} \) is about \( 10^{-3} \).

Through the weak interaction, the close-lying opposite-parity levels admix each other. The eigenstates of the levels no longer have a well-defined parity. According to the perturbation theory, new eigenstate \( |f'\rangle \) is expressed as

\[
|f'\rangle = |f^+\rangle \pm \delta_{PNC} |n^-\rangle,
\]

where \( \delta_{PNC} = \frac{\langle n^- | V_{PNC} | f^+ \rangle}{\Delta E} \).

The PNC mixing coefficient is denoted by \( \delta_{PNC} \). The denominator \( \Delta E \) is the energy separation between the levels \( |f^+\rangle \) and \( |n^-\rangle \). The size of \( \delta_{PNC} \) is of the order of \( 10^{-9} \). The sign \( \pm \) expresses
CHAPTER 2. THEORETICAL DESCRIPTIONS

the handedness of the experimental coordinate system. As a result, the electric dipole \((E_{1\text{PNC}})\) transition is induced between the levels \(|i^+\rangle\) and \(|f\rangle\) by the PNC effect. The \(E_{1\text{PNC}}\) transition amplitude is written as

\[
A_{\text{PNC}} = \delta_{\text{PNC}} A_{E_1} \propto Q_W. \tag{2.11}
\]

Here we ignore the PNC mixing contributing to the level \(|i^+\rangle\) since the energy separation between the levels \(|i^+\rangle\) and \(|n^-\rangle\) is relatively large.

As the \(E_{1\text{PNC}}\) transition amplitude is extremely small, it is difficult to observe the \(E_{1\text{PNC}}\) transition directly. In the Stark-interference method, external electric field is applied to the atoms to induce an additional parity mixing between the levels \(|f^+\rangle\) and \(|n^-\rangle\);

\[
|\overline{f}\rangle = |f^+\rangle \pm \delta_{\text{Stark}} |n^-\rangle + \delta_{\text{Stark}} |n^-\rangle, \tag{2.12}
\]

where

\[
\delta_{\text{Stark}} = \frac{\langle n^-|H_{\text{Stark}}|f^+\rangle}{\Delta E}. \tag{2.13}
\]

\(H_{\text{Stark}}\) is the Stark interaction Hamiltonian: \(H_{\text{Stark}} = -E \cdot d\), where \(E\) and \(d\) are the electric field vector and atomic electric dipole operator, respectively.

The electric dipole \((E_{1\text{Stark}})\) transition is induced by the electric filed between the levels \(|i^+\rangle\) and \(|\overline{f}\rangle\) with the transition amplitude written as

\[
A_{\text{Stark}} = \delta_{\text{Stark}} A_{E_1} \propto E. \tag{2.14}
\]

Its size is proportional to the applied electric field strength \(E\).

In this system, the transition intensity between the levels \(|i^+\rangle\) and \(|\overline{f}\rangle\) is written as

\[
I_{\pm} = |A_{M1} \pm A_{\text{PNC}} + A_{\text{Stark}}|^2 = |A_{\text{Stark}}|^2 \pm 2\text{Re}(A_{\text{Stark}} A_{\text{PNC}}^*) + 2\text{Re}(A_{\text{Stark}} A_{M1}^*) + \cdots. \tag{2.15}
\]

The second term, which is the interference between \(A_{\text{Stark}}\) and \(A_{\text{PNC}}\), yields the asymmetry in the transition with the reversal of the handedness. The third term, which is the interference between \(A_{\text{Stark}}\) and \(A_{M1}\), is canceled out by choosing a specific experimental condition such that the laser field is the standing-wave of circular-polarized light [19]. The magnetic field is also applied to the atom in order to define the handedness of the coordinate system.

In the experiment, the asymmetry in the transition intensity is measured as follows,

\[
P = \frac{|I_+ - I_-|}{I_+ + I_-} \simeq \frac{2\text{Re}(A_{\text{Stark}} A_{\text{PNC}}^*)}{|A_{\text{Stark}}|^2} \propto \frac{Q_W}{E}, \tag{2.16}
\]

when the handedness is reversed. To extract \(Q_W\) from the measured asymmetry, the calculation of matrix elements \(\langle n^-|H_{\text{Stark}}|f^+\rangle\) and \(\langle n^-|V_{\text{PNC}}|f^+\rangle\) are required. In the calculation, the ambiguity of wave function can be canceled out by taking the ratio between different isotopes.

#### 2.1.3 Sensitivity to New Physics beyond Standard Model

The electroweak observable obtained in the atomic PNC experiment can play a special role for the tests of the standard theory. In the weak-interaction processes involving only light quarks and leptons, the effects of new physics beyond the standard model appear through vacuum polarization of the photon, \(Z\) and \(W\) bosons (so called “oblique corrections”). This oblique corrections have been parametrized by Peskin and Takeuchi in terms of isospin-conserving \(S\) and -breaking \(T\) [20].
2.1. ATOMIC PARITY NON-CONSERVATION

Table 2.1: Electroweak observables. Theoretical values are given as function of $S$ and $T$ (tabulated by Rosner [13]).

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Exp. value</th>
<th>Theo. value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Q_W$(Cs)</td>
<td>$-72.2 \pm 0.8^{(a)}$</td>
<td>$-73.19^{(b)} - 0.800S - 0.007T$</td>
</tr>
<tr>
<td>$Q_W$(Tl)</td>
<td>$-115.0 \pm 4.5^{(c)}$</td>
<td>$-116.8^{(d)} - 1.17S - 0.06T$</td>
</tr>
<tr>
<td>$M_W$(GeV/c$^2$)</td>
<td>$80.451 \pm 0.033^{(e)}$</td>
<td>$80.385^{(f)} - 0.29S - 0.45T$</td>
</tr>
<tr>
<td>$\Gamma_\alpha(Z)$ (MeV)</td>
<td>$83.991 \pm 0.087^{(g)}$</td>
<td>$84.011^{(f)} - 0.18S - 0.78T$</td>
</tr>
<tr>
<td>$\sin^2 \theta_{\text{eff}}$</td>
<td>$0.23152 \pm 0.00017^{(h)}$</td>
<td>$0.23140^{(f)} - 0.00362S - 0.00258T$</td>
</tr>
<tr>
<td>$\mu^2(W)$ (GeV/c$^2$)</td>
<td>$80.136 \pm 0.084^{(i)}$</td>
<td>$80.385^{(f)} - 0.27S - 0.56T$</td>
</tr>
</tbody>
</table>

(d) Calculation incorporating electroweak corrections $[29]$.
(h) NuTeV value from deep inelastic neutrino scattering $[33]$.

The $W^\pm$ and $Z^0$ boson self-energies at momentum transfer $q$, $\Pi_{WW}(q^2)$ and $\Pi_{ZZ}(q^2)$, contribute to their propagators $[3]$:

$$\frac{1}{q^2 - M_{W}^0 - \Pi_{WW}(q^2)}, \quad \frac{1}{q^2 - M_{Z}^0 - \Pi_{ZZ}(q^2)}.$$  \hfill (2.17)

where $M_{W}^0$ and $M_{Z}^0$ are the bare masses of $W^\pm$ and $Z^0$ bosons, respectively. Assuming that all the standard-model loop corrections are properly known, we consider only the new-physics contributions $\Pi_{WW}(q^2)$ and $\Pi_{ZZ}(q^2)$. The parameter $S$ comes from a momentum-transfer dependence of neutral current processes;

$$\left[ \frac{\Pi_{WW}(M_{Z}^2) - \Pi_{WW}(0)}{M_{Z}^2} \right]_{\overline{\text{MS}}} = \frac{\alpha(M_{Z})}{4\cos^2 \theta_{\text{W}}(M_{Z})_{\overline{\text{MS}}}} S,$$  \hfill (2.18)

where $\alpha(M_{Z}) \simeq 1/128.933 \pm 0.021 \ [21]$ and $\cos^2 \theta_{\text{W}}(M_{Z})_{\overline{\text{MS}}}$ are given in the mass scale of $M_{Z}$. The subscript $\overline{\text{MS}}$ denotes the modified minimal subtraction $[22]$. The parameter $T$ comes from the difference of new-physics contribution between charged and neutral current processes at $q^2 = 0$;

$$\rho(0)^{\text{new}} = 1 + \left[ \frac{\Pi_{WW}(0)}{M_{W}^2} - \frac{\Pi_{ZZ}(0)}{M_{Z}^2} \right] = 1 + \alpha(M_{Z})T.$$  \hfill (2.19)

The electroweak observables are expressed as linear functions of $S$ and $T$ with precise electroweak parameters $G_{F} = 1.16637(1) \times 10^{-5} \ \text{GeV}^{-2}$, $\alpha^{-1}(M_{Z}) = 128.933 \pm 0.021$ and $M_{Z} = 91.1874 \pm 0.0021 \ \text{Gev}/c^2$. Thus precise measurements of electroweak observables can provide an allowed band in the $S-T$ plane. The point where $S \neq 0$ or $T \neq 0$ indicates the existence of “new physics”.

Rosner has recently reviewed the precision electroweak tests $[13]$ including several reanalysis of Cs. Various measured electroweak observables are listed in Table 2.1 accompanying with the theoretical values expressed with $S$ and $T$. The experimental values of $Q_W$(Cs) and $Q_W$(Tl) are derived in the PNC measurements. The others are derived in high-energy experiment. As shown
in the Table, $Q_W$ is sensitive to $S$ and small dependence on $T$ while the dependence of the others on $S$ and $T$ are comparable. Thus the allowed bands obtained in the PNC measurements give strong constraint on “new physics” by combining those obtained in high-energy experiments.

The most accurate value of $Q_W$ in Cs has been reported by JILA-Boulder group [5,12]. Their resulting value was $Q_W(Cs) = -72.06 \pm 0.28_{\text{expt}} \pm 0.34_{\text{theor}} = -72.06 \pm 0.46$ [12], which was 2.3 standard deviations away from the standard model prediction $Q_W(Cs) = -73.19 \pm 0.13$ [3,23]. The re-evaluations of $Q_W$ have been done by including the Breit interaction [24–27]. Rosner has presented the averaged value of $Q_W(Cs) = -72.2 \pm 0.8$ [13]. It indicates no remarkable deviation from the standard model.

In Fig. 2.3, the allowed range of $S$ and $T$ is shown, which is obtained from the present constraints in Table 2.1. The diagonal bands bounded by dashed lines correspond to $\pm 1\sigma$ components associated with direct $M_W$ measurements and with NuTeV measurements [33]. A banana-shape grid represents minimal standard model with top quark mass $m_t = 174 \pm 5.1$ GeV and Higgs boson mass $M_H$ running from 100 to 1000 GeV. The fits with and without the PNC data are compared in Table 2.2.

As another aspect of the sensitivity to new physics, it has been pointed that the electric dipole moment (EDM) on the electron is enhanced in Sm, where the enhancement factor is evaluated to be in range of 100–3800 [34]. The measurement of EDM can directly show the existence of new physics, that is a time-reversal (T) asymmetry [35].
2.2 Electromagnetic Interactions in Atom

2.2.1 Isotope Shift

In a transition \( i \), the observed isotope shift \( \delta \nu_i \) between two isotopes with mass numbers \( A \) and \( A' \) is expressed as the sum of normal mass shift (NMS), specific mass shift (SMS) and field shift (FS) \([36,37]\),

\[
\delta \nu_i = \delta \nu_{\text{NMS}} + \delta \nu_{\text{SMS}} + \delta \nu_{\text{FS}}. \tag{2.20}
\]

The NMS is caused by the reduced-mass correction between an orbital electron and the nucleus in an atom. The SMS is caused by the correlation between orbital electrons. The NMS and the SMS are written as

\[
\delta \nu_{\text{NMS}} = M_{\text{NMS}} \frac{A' - A}{AA'}, \tag{2.21}
\]

\[
\delta \nu_{\text{SMS}} = M_{\text{SMS}} \frac{A' - A}{AA'}, \tag{2.22}
\]

where \( M_{\text{NMS}} \) and \( M_{\text{SMS}} \) are factors of NMS and SMS, respectively. The \( M_{\text{NMS}} \) has the simple expression,

\[
M_{\text{NMS}} = \frac{m_e}{m_p} \nu_i, \tag{2.23}
\]

where \( m_p \) is the proton mass, and \( \nu_i \) is the transition frequency. In case of a pure \( s^2 - sp \) transition, the SMS is negligibly small, and the semiempirical evaluation is used as

\[
\delta \nu_{\text{SMS}} = (0 \pm 0.5) \delta \nu_{\text{NMS}}. \tag{2.24}
\]

The FS, which comes from the change in nuclear charge distribution, is written as

\[
\delta \nu_{\text{FS}} = E_i \cdot f(Z) \cdot \lambda = \frac{\pi a_0^3}{Z} \Delta |\psi(0)|^2 \cdot f(Z) \cdot \lambda, \tag{2.25}
\]

where \( a_0 \) is the Bohr radius, and \( \Delta |\psi(0)|^2 \) is the electron density difference at the nucleus between the lower and upper electronic levels for the transition \( i \). A relativistic correction factor \( f(Z) \) is a function of the atomic number \( Z \). The nuclear parameter \( \lambda \) is related to changes in mean square nuclear charge radii and higher order contributions.

2.2.2 Hyperfine Structure

The interaction between the orbital electrons and the nucleus yields the coupling of the nuclear spin \( I \) and the electronic angular momentum \( J \), so called hyperfine interaction characterized by the atomic total angular momentum \( \mathbf{F} \):

\[
\mathbf{F} = \mathbf{I} + \mathbf{J}. \tag{2.26}
\]

The hyperfine Hamiltonian is written as \([38]\)

\[
H_{\text{hfs}} = A_{\text{hfs}} \mathbf{I} \cdot \mathbf{J} + B_{\text{hfs}} \frac{3(\mathbf{I} \cdot \mathbf{J})^2 + (3/2)(\mathbf{I} \cdot \mathbf{J}) - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}. \tag{2.27}
\]

The parameters \( A_{\text{hfs}} \) and \( B_{\text{hfs}} \) are the hyperfine coupling constants;

\[
A_{\text{hfs}} = \frac{\mu_{I} \overline{P}(0)}{IJ}, \tag{2.28}
\]

\[
B_{\text{hfs}} = eQ_{I} \overline{\varphi}_{jj}(0), \tag{2.29}
\]
where $e$ is elementary charge, $\mu_I$ and $Q_I$ are the magnetic dipole and the electric quadrupole moments of the nucleus, respectively. $\overrightarrow{H}(0)$ is the magnetic field, $\overrightarrow{\nabla}j(0)$ is the electric field gradient generated by the electron cloud at the site of the nucleus. Diagonalizing $H_{\text{hfs}}$, the splitting energy is written as

$$\Delta E_{\text{hfs}} = \frac{1}{2} K(F) A_{\text{hfs}} + L(F) B_{\text{hfs}},$$

$K(F) = F(F + 1) - I(I + 1) - J(J + 1)$,

$L(F) = \frac{(3/4)K(K + 1) - I(I + 1)J(J + 1)}{2I(2I - 1)J(2J - 1)}$.

### 2.2.3 Stark Effect

The electric field strength available in the laboratory is of the order of $10^6$ V/cm, which is far small compared with the atomic Coulomb field;

$$\mathcal{E}_{\text{Coulomb}} = \frac{e}{4\pi\varepsilon_0\alpha_0^3} = 5 \times 10^9\text{V/cm}.$$ 

Thus the Stark effect is treated within a perturbation theory. According to the time-independent perturbation theory, the eigenvalue $W$ and the eigenstate $|\psi\rangle$ can be written up to the second order as \[39\]

$$W = E_m + \langle m | H_{\text{Stark}} | m \rangle + \sum_{n \not= m} \langle m | H_{\text{Stark}} | n \rangle \frac{|\langle m | H_{\text{Stark}} | m \rangle|^2}{E_m - E_n},$$

$$|\psi\rangle = |m\rangle + \sum_{k \not= m} |k\rangle \frac{\langle k | H_{\text{Stark}} | m \rangle}{E_m - E_k}$$

$$+ \sum_{k \not= m} |k\rangle \left[ \sum_{n \not= m} \frac{\langle k | H_{\text{Stark}} | n \rangle \langle n | H_{\text{Stark}} | m \rangle}{(E_m - E_k)(E_m - E_n)} - \frac{\langle k | H_{\text{Stark}} | m \rangle \langle m | H_{\text{Stark}} | m \rangle}{(E_m - E_k)^2} \right],$$

where $E_{m,n,k}$ are the unperturbed eigenvalues of the states $|m\rangle$, $|n\rangle$, $|k\rangle$, respectively. The perturbation Hamiltonian $H_{\text{Stark}}$ for the external electric field can be written in the dipole approximation as

$$H_{\text{Stark}} = -\mathcal{E} \cdot \mathbf{d},$$

$$\mathbf{d} = -e \sum_i r_i.$$ 

Since the perturbation Hamiltonian $H_{\text{Stark}}$ does not commute with the parity operator but the anticommutator vanishes, the matrix elements of $H_{\text{Stark}}$ between the states of opposite parity only exist. Then the first-order Stark effect in Eq. (2.31) does not exist except in the case such that the states of opposite parity are degenerate as in a hydrogen atom. Here we take the direction of $\mathcal{E}$ along $z$ axis, Stark Hamiltonian is written as $H_{\text{Stark}} = -\mathcal{E}d_z$, where $\mathcal{E}$ is the electric field strength, and $d_z$ is the $z$ component of $d$. Thus the Stark shift of the state $|nJm\rangle$ is written as

$$\Delta E_{\text{Stark}}(nJm) = \sum_{n'Jm'} \frac{|\langle nJm | H_{\text{Stark}} | n'Jm' \rangle|^2}{E_{nJ} - E_{n'J'}}$$

$$= \mathcal{E}^2 \sum_{n'J'} \frac{|\langle nJm | d_z | n'Jm' \rangle|^2}{E_{nJ} - E_{n'J'}}.$$

### References

\[39\]
where \( n, J \) and \( m \) denote the principal quantum number, the total angular momentum and the projection of \( J \) on the quantization axis, respectively. Using the Wigner-Eckart theorem \([40]\) together with \( 3\)-\( j \) symbol, we have

\[
\langle nJm|d_z|n'J'm' \rangle = (-1)^{J-m} \begin{pmatrix} J & 1 & J' \\ m & 0 & -m' \end{pmatrix} \langle nJ|d|n'J' \rangle
\]

\[
\langle nJ|d|n'J' \rangle = \begin{cases} 
\sqrt{J^2 - m^2} \quad & \text{for } J' = J - 1, m' = m \\
\frac{m}{\sqrt{J(J+1)(2J+1)}} \quad & \text{for } J' = J, m' = m \\
\frac{\sqrt{(J+1)^2 - m^2}}{\sqrt{(J+1)(2J+1)(2J+3)}} \quad & \text{for } J' = J + 1, m' = m \\
0 & \text{for } |J' - J| > 1 \text{ or } m' \neq m \text{ or } J = J' = 0.
\end{cases}
\]

In terms of scalar and tensor polarizabilities \( (\alpha_0 \text{ and } \alpha_2) \), the Stark shift is written in the form of

\[
\Delta E_{\text{Stark}}(nJm) = -\frac{1}{2} \alpha_0 \epsilon^2 - \frac{1}{2} \alpha_2 \frac{3m^2 - J(J+1)}{J(2J-1)} \epsilon^2,
\]

where

\[
\alpha_0 = -\frac{2}{3(2J+1)} \sum_{J'} \frac{|\langle Jd|J' \rangle|^2}{E_{J'} - E_J},
\]

\[
\alpha_2 = 2 \left[ \frac{10J(2J-1)}{3(2J+3)(J+1)(2J+1)} \right]^{1/2} \sum_{J'} (-1)^{J+J'+1} \times \begin{pmatrix} J & 1 & 2 \\ J' & 1 & 1 \end{pmatrix} \frac{|\langle Jd|J' \rangle|^2}{E_{J'} - E_J}.
\]

Assuming that the Stark mixing is dominated with one level, which is the closest lying neighbor, the tensor polarizability is written as

\[
\alpha_2 = \begin{cases} 
\frac{2}{3} \frac{J(2J-1)}{(E_{nJ} - E_{n'J})} \frac{|\langle nJ|d|n'J' \rangle|^2}{J(J+1)(2J-1)} \quad & \text{for } J' = J - 1 \\
\frac{2}{3} \frac{J(2J-1)}{(E_{nJ} - E_{n'J})} \frac{|\langle nJ|d|n'J' \rangle|^2}{J(J+1)(2J+1)} \quad & \text{for } J' = J \\
2 \frac{J(2J-1)}{(E_{nJ} - E_{n'J})} \frac{|\langle nJ|d|n'J' \rangle|^2}{(J+1)(2J+1)(2J+3)} \quad & \text{for } J' = J + 1.
\end{cases}
\]

The magnitude of dipole matrix element is expressed by using \( \alpha_2 \):

\[
|\langle nJm|d_z|n'J'm' \rangle| = \begin{cases} 
\sqrt{\frac{3(J^2 - m^2)}{2J(2J-1)}} \alpha_2(E_{nJ} - E_{n'J}) \quad & \text{for } J' = J - 1 \\
\sqrt{\frac{3m^2}{2J(2J-1)}} \alpha_2(E_{nJ} - E_{n'J}) \quad & \text{for } J' = J \\
\frac{3((J+1)^2 - m^2)}{2J(2J-1)} \alpha_2(E_{nJ} - E_{n'J}) \quad & \text{for } J' = J + 1.
\end{cases}
\]
Maximum magnitude of dipole matrix element is given as

\[
|\langle nJm|dz|n'J'm'\rangle|_{\text{max}} = \begin{cases} 
\sqrt{\frac{3}{2(2J-1)}} \alpha_2 (E_{nJ} - E_{n'J'}) & \text{for } J' = J - 1, \ m = 0 \\
\sqrt{\frac{3}{2(2J-1)}} \alpha_2 (E_{nJ} - E_{n'J'}) & \text{for } J' = J, \ m = \pm J \\
\sqrt{\frac{3}{2J(2J-1)}} (J + 1)^2 \alpha_2 (E_{nJ} - E_{n'J'}) & \text{for } J' = J + 1, \ m = 0 
\end{cases}
\] (2.41)

2.2.4 Zeeman Effect

In case of a fine structure level specified by \( J \), the interaction Hamiltonian of the atom under the external magnetic field is written as \[41\]

\[H_{\text{Zeeman}} = \mu_B g_J J_Z B,\] (2.42)

where \( \mu_B \) is the Bohr magneton, \( g_J \) is the Lande \( g \)-value and \( B \) is the magnetic flux density. \( J_Z \) is the projection of the electronic total angular momentum on the quantization axis. For sufficiently small values of \( B \), the energy under the external magnetic field is calculated in simple perturbation treatment. Diagonalizing \( H_B \), the \( (2J+1) \)-fold degeneracy of each level is completely removed, the fine structure level splits into a set of \( 2J+1 \) sub-levels. The energy shift of each sub-level is written as

\[\Delta E_{\text{Zeeman}} = \mu_B g_J m_J B,\] (2.43)

where \( m_J \) is the magnetic quantum number. When the external magnetic field is sufficiently strong, the effect of \( B \) is enough to decouple \( L \) from \( S \) in case of near LS coupling and to make \( J \) meaningless, which is called Paschen-Back effect. In place of Eq. (2.43), we have

\[\Delta E_{\text{Zeeman}} = \mu_B (m_L + g_S m_S) B,\] (2.44)

where \( g_S \) is a free electron \( g \)-value.
Chapter 3
PNC Measurement in Atomic Samarium

3.1 Candidates for the PNC Measurements in Sm

3.1.1 Spectroscopic Properties of Atomic Samarium

The rare-earths consist of two series of elements, the lanthanides ($Z=57–70$) and the actinides ($Z=89–102$), which, roughly speaking, involve filling of $4f$ and $5f$ subshells, respectively. The unique properties of the lanthanide elements are a direct consequence of the small radii of the $4f$ orbitals. The $4f$ electrons are well shielded from outer valence electrons and environmental effects, and their binding energies are relatively small and comparable with those of $5d$, $6s$ and $6p$ electrons.

Atomic samarium belongs to the lanthanides having atomic number of 62 and atomic weight of 150.36. The melting and boiling points are 1353 K and 2073 K, respectively. The naturally occurring abundance and the nuclear spin for each isotope are listed in Table 3.1. Although $^{144}$Sm is a radio active isotope, its life is extremely long.

The lowest configuration of Sm is an inner shell of xenon (Xe) plus valence electrons of $4f^66s^2$:

$$\text{(Xe)}4f^66s^2 = (1s^22s^22p^63s^23p^63d^{10}4s^24p^64d^{10}5s^25p^6)4f^66s^2^7F_0.$$ 

The identified even configurations are $4f^66s^2$, $4f^65d6s$ and $4f^66s7s$. The identified odd configurations are $4f^66s6p$, $4f^55d6s^2$, $4f^55d^26s^2$ and $4f^65d6p$. About 500 energy level data have been tabulated by Martin et al. [42]. It includes the information about the electronic configuration, term, parity, $J$ and $g$-value.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$^{144}$Sm*</th>
<th>$^{147}$Sm</th>
<th>$^{148}$Sm</th>
<th>$^{149}$Sm</th>
<th>$^{150}$Sm</th>
<th>$^{152}$Sm</th>
<th>$^{154}$Sm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nuclear spin</td>
<td>0</td>
<td>7/2</td>
<td>0</td>
<td>7/2</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>
CHAPTER 3. PNC MEASUREMENT IN ATOMIC SAMARIUM

Table 3.2: Calculated excitation energy of the low-lying levels in cm\(^{-1}\). MCDF: multi-configuration Dirac-Fock method. CI1: method of configuration interaction using the orbitals generated so far. CI2: method of configuration interaction after including configuration state functions. The abbreviation Config. denotes the configuration.

<table>
<thead>
<tr>
<th>Config. and J</th>
<th>Expt.</th>
<th>Porsev(^a)</th>
<th>Present work</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>MCDF</td>
</tr>
<tr>
<td>4f(^6)6s(^2)7F(_1)</td>
<td>292.58</td>
<td>285</td>
<td>276</td>
</tr>
<tr>
<td>4f(^6)6s(^2)7F(_2)</td>
<td>811.92</td>
<td>810</td>
<td>790</td>
</tr>
<tr>
<td>4f(^6)6s(^6)p(^9)G(_0)</td>
<td>13796.36</td>
<td>11339</td>
<td>11147</td>
</tr>
<tr>
<td>4f(^6)6s(^6)p(^9)G(_1)</td>
<td>13999.50</td>
<td>11533</td>
<td>11337</td>
</tr>
<tr>
<td>4f(^6)6s(^6)p(^9)G(_3)</td>
<td>14380.50</td>
<td>11906</td>
<td>11704</td>
</tr>
<tr>
<td>4f(^5)5d(^6)s(^9)H(_1)</td>
<td>10801.10</td>
<td>12739</td>
<td>13227</td>
</tr>
<tr>
<td>4f(^5)5d(^6)s(^9)H(_2)</td>
<td>11044.90</td>
<td>12693</td>
<td>13444</td>
</tr>
</tbody>
</table>

\(^a\) Ref. [51].

The spectroscopic studies were performed for the ground term of 4f\(^6\)6s\(^2\)7F. The hyperfine coupling constants and g-values have been precisely determined by Childs et al. [43], and the tensor polarizabilities \(\alpha_2\) have been determined by Rinkleff et al. [44]. A large number of optical transitions exist in the visible and near-infrared regions owing to the complicated electronic structures. Many studies of the hyperfine structure and isotope shift for low-lying odd-parity levels have been reported [36,37,45]. The spectroscopic studies for high-lying odd-parity levels have been carried out by means of the electric discharge method [46–48]. In contrast, the studies for the low-lying even-parity levels are very few. Three 4f\(^6\)6s\(^2\)5D levels, which are not listed in Ref. [42], have been identified by Barkov et al. [49].

3.1.2 Theoretical Calculations of Sm

Obtaining the precise \(Q_W\) requires atomic wave-functions which are accurate at all radial ranges. Thus it is important to check the accuracy of the wave-function by comparing the theoretically calculated atomic properties with the experimental data. The excitation energies and hyperfine coupling constants of the low-lying levels have been calculated using the multi-configuration Dirac-Fock (MCDF) method [50]. The calculated excitation energy of the low-lying levels is shown in Table 3.2. The calculated hyperfine coupling constants of 4f\(^6\)6s\(^2\)7F\(_1\) level for \(^{147}\)Sm is shown in Table 3.3. These results show that the excitation energies of the ground term can be calculated accurately using a small orbital set but the higher levels need larger orbital sets. Similar interface can also be drawn from the discrepancy between the calculated values of the hyperfine coupling constants and the experimental data.

3.1.3 Feasible Levels for Measuring the PNC Effects

Among the rare-earth elements, the combinations of levels suited for the PNC measurements were searched using the database of rare-earth atomic energy levels [53,54]. The pairs of opposite-parity fine-structure levels with the same electronic angular momentum \(J\) were searched for, and many pairs having small energy separations were found. The close-lying opposite-parity pairs in Sm, whose energy separation \(\Delta E\) are less than 30 cm\(^{-1}\), are listed in Table 3.4. Taking into
### Table 3.3: Calculated hyperfine coupling constants of $4f^66s^27F_1$ level for $^{147}$Sm in units of MHz. CSF denotes the configuration state function. $n_c$ represents the number of CSFs used in the calculation.

<table>
<thead>
<tr>
<th>CSFs</th>
<th>$n_c$</th>
<th>$A_{hfs}$</th>
<th>$B_{hfs}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$6s^2 + 5d6s + 5d^2$</td>
<td>5540</td>
<td>-16.88</td>
<td>-36.34</td>
</tr>
<tr>
<td>+6$p^2$</td>
<td>6974</td>
<td>-16.81</td>
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<tr>
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<tr>
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<tr>
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<td></td>
<td>-33.493</td>
<td>-58.688</td>
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$^a$) Ref. [52]. $^b$) Ref. [43].

### Table 3.4: Close-lying opposite-parity pair and $\alpha_2$ of odd-parity level in Sm. The pairs whose energy separation $\Delta E$ is less than 30 cm$^{-1}$ are listed. The levels of the pairs have the same $J$. The abbreviation Config. denotes the configuration. Energy and $\Delta E$ are given in units of cm$^{-1}$. $\alpha_2$ is given in units of kHz/(kV/cm)$^2$. NR: the Stark splitting is not resolved. NM: not measured.

<table>
<thead>
<tr>
<th>Even-parity level</th>
<th>Energy</th>
<th>Config.</th>
<th>Level</th>
<th>Odd-parity level</th>
<th>Energy</th>
<th>Config.</th>
<th>Level</th>
<th>$\alpha_2$</th>
<th>$\Delta E$</th>
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<td>14026.45</td>
<td>$4f^65d6s$</td>
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<td>13999.50</td>
<td>$4f^66s6p$</td>
<td>$9G_1$</td>
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<td></td>
<td>14365.50</td>
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<td></td>
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<td>$9F_3$</td>
<td>28.4(10)$^a$</td>
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<td>$J = 1$</td>
<td>NR$^c$</td>
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</table>

CHAPTER 3. PNC MEASUREMENT IN ATOMIC SAMARIUM

account the PNC measurement with Stark-interference method, the tensor polarizabilities for odd-parity levels are listed.

Several groups are especially interested in the pair of $15639.80 \text{ cm}^{-1}$ ($4f^66d6s^7G_1$) and $15650.55 \text{ cm}^{-1}$ ($4f^66s6p^7G_0$) levels, and the Stark effect has been precisely studied for the $15650.55 \text{ cm}^{-1}$ level [34, 56, 57]. It found that the value of $\alpha_2$ is relatively large. The opposite-parity pair is also a candidate for studying the time-reversal violations, that is the measurement of an atomic electric dipole moment [34].

3.1.4 Estimation of Asymmetry

The PNC mixing matrix element between the $15650.55 \text{ cm}^{-1}$ and $15639.80 \text{ cm}^{-1}$ levels has been calculated by Gongora and Sandars as [15]

$$|\langle 15650.55 | V_{\text{PNC}} | 15639.80 \rangle| \simeq 3 \times 10^{-13} \text{ Ry},$$

(3.1)

where atomic energy is expressed in units of the Rydberg (Ry). The Rydberg for an atom having nuclear mass $M$ is

$$1 \text{ Ry} = \frac{M}{M + m_e} R_\infty,$$

(3.2)

where $R_\infty$ is a Rydberg constant. Substituting the energy separation of $10.75 \text{ cm}^{-1}$ together with Eq. (3.1) to Eq. (2.10) yields the PNC mixing coefficient as

$$\delta_{\text{PNC}} = 3.1 \times 10^{-9}.$$  

(3.3)

On the other hand, the Stark mixing coefficient for the pair is estimated by using measured $\alpha_2$ [34];

$$\delta_{\text{Stark}} \simeq 1.6 \times 10^{-3} \text{ cm/kV } \mathcal{E}.$$ 

(3.4)

Using Eq. (3.3) and Eq. (3.4), the asymmetry in the transition intensity is estimated to be

$$P \simeq \frac{2 \text{Re}(A_{\text{Stark}} A_{\text{PNC}}^*)}{|A_{\text{Stark}}|^2} = \frac{2 \delta_{\text{PNC}}}{\delta_{\text{Stark}}} \equiv \frac{p}{\mathcal{E}},$$ 

(3.5)

with

$$p \simeq 3.9 \times 10^{-6} \text{ kV/cm}.$$ 

(3.6)

3.2 Two-step Excitation System for PNC Experiments

3.2.1 Previous PNC Measurement in Sm

The PNC measurement using Sm was first discussed by Gongora and Sandars [15]. They have suggested the feasibility of the PNC measurement with optical rotation method. The experimental studies with the vapor cell have been performed to investigate possible large enhancement [16, 17]. They have searched for parity nonconserving optical rotation in several M1 transitions and concluded that Sm transitions did not reveal strong PNC effects enough to offer a critical test of the standard theory. The possible reasons for the absence of the enhancement comes from two contributory factors: the matrix elements of $E1$ amplitude and $H_{\text{PNC}}$ were so small that they might have offset the potential gain given by the energy near-degeneracy.

However, for the parity forbidden transition from the $0 \text{ cm}^{-1}$ to $15639.80 \text{ cm}^{-1}$ levels, Rochester et al. has estimated the $E1_{\text{PNC}}$ amplitude to be $4 \times 10^{-6} e a_0$ [34] based on an
3.2. TWO-STEP EXCITATION SYSTEM FOR PNC EXPERIMENTS

experimental value of the $E1$ amplitude measurement. The $E1_{\text{PNC}}$ amplitude is comparable with those for other elements, such as ytterbium ($\sim 10^{-9}a_0$) and cesium ($\sim 10^{-11}a_0$).

Although the investigation in Sm was already done with the optical rotation method, the experiments based on the fluorescence detection have not been performed yet. It is still worth while studying Sm with another experimental method in which we can solve the problem offsetting the potential gain.

3.2.2 Suggestion of Two-step Excitation System

In the experiment with the Stark interference method, the simplest experimental scheme is to detect the fluorescence after exciting the parity forbidden transition from the ground level to low-lying even-parity level which has close-lying neighbor as shown in Fig. 3.1 (a). However, Kuwamoto pointed out that there is a lifetime problem in Sm level structure [48]. After exciting the 15639.80-cm$^{-1}$ level, the atoms decay via two-step $E1$ processes. As the first decay transition lies around middle and far infrared region, the 15639.80-cm$^{-1}$ level is regarded to be long-lived. Its lifetime was estimated to be $\sim 70\mu s$ [48] and $\sim 1\text{ ms}$ [34], where the discrepancy between two estimations came from the difference of the assumption of typical $E1$ matrix elements. In the experimental system based upon the atomic-beam laser spectroscopy, the detection efficiency might decrease for detecting the fluorescence from the long-lived level.

A two-step excitation system with optical double-resonance (ODR) technique was proposed as shown in Fig. 3.1 (b) [48]. Using two lasers, atoms are finally excited to a high-lying odd-parity level around 30000 cm$^{-1}$. As the lifetime of the levels around 30000 cm$^{-1}$ is very short (typically 10–100 ns [58]), the atoms immediately decay from the level to the ground or metastable levels. Moreover, as the wavelength of the fluorescence is quite different from those of excitation lasers, it is possible to block the background caused by the laser scattered light with a specific optical filter.

The two-step excitation system has another advantage. As the smallness of $E1$ amplitude seems to have prevented an observed enhancement of PNC effects in the previous studies, we may still expect the enhancement in Sm with an experimental scheme where a larger $E1$ amplitude contributes to the $E1_{\text{PNC}}$ amplitude. An optical double-resonance technique would enable us to...
search for a favorable transition in a wide energy range. In an atomic level scheme as shown in Fig. 3.2, the 15650.55-cm$^{-1}$ level, which is the closest opposite parity neighbor to the 15639.80-cm$^{-1}$ level, is populated by a pump laser. A probe laser is tuned to the parity forbidden transition from the 15650.55-cm$^{-1}$ level to a high-lying odd-parity level $X$. In the scheme, if the large transition amplitude from the 15639.80-cm$^{-1}$ level to level $X$ is found, the transition from the 15650.55-cm$^{-1}$ to level $X$ is a strong candidate for the PNC measurement.

Because of the large tensor polarizability of the 15650.55-cm$^{-1}$ level [34], a Stark-interference method with an atomic-beam of Sm could be used. We start here with a measurement to observe the Stark-induced $E1$ transition from the 15650.55-cm$^{-1}$ to level $X$ via Stark mixing with the 15639.80-cm$^{-1}$ level.
Chapter 4

Overview of Experiments

4.1 Experimental Procedure

Our experimental procedure consists of three stages as

- spectroscopic study of low-lying levels,
- exploration of high-lying levels, and
- measurement of the Stark-induced $E_1$ transition.

The first stage is spectroscopic study of $4f^66s6p^7G$ term. Our interest especially lies in the $15650.55$-cm$^{-1}$ ($4f^66s6p^7G_1$) level which is expected the large parity mixing caused by the close-lying opposite-parity neighbor. In order to perform the PNC measurements, it is necessary to investigate the spectroscopic properties, such as isotope shift, hyperfine structure, Stark effect and Zeeman effect. We systematically perform the spectroscopic study of $4f^66s6p^7G$ term. The second stage is to explore the second excited levels which is suitable for the optical double-resonance. The investigation of such high-lying levels is carried out with electric discharge method. The electric discharge method populates the metastable levels and enables us to measure the high-lying levels with one laser. The final stage is the observation of Stark-induced $E_1$ transition, which is the purpose of this study.

The basic apparatus which is common to all experimental stages is described in Section 4.2. We also introduce the laser-frequency locking system constructed for the optical double-resonance experiments in Section 4.3.

4.2 Apparatus

4.2.1 Vacuum System

The schematic drawing of the vacuum chamber is shown in Fig. 4.1. The vacuum chamber is evacuated with two turbo molecular pump systems (OSAKA VAC. TF-160 + ULVAC GVD-050A and UVLVAC UTM-500 + ULVAC VD301). The chamber has a cylindrical shape with about 1-m length and 21-cm inner diameter. The laser ports are attached to the chamber. The inside of the chamber consists of two rooms. One is for atomic beam generation, and the other is for interactions with lasers. They are separated by a partition board with the slits for the
Figure 4.1: Schematic drawing of vacuum chamber. PMT represents a photomultiplier tube. IP1 and IP2 denote upstream and downstream interaction points, respectively. The symbols \( \mathbf{k} \), \( \mathbf{E} \) and \( \mathbf{B} \) represent the directions of the incident laser, electric field and magnetic field, respectively.
atomic beam. When the atomic beam is produced from the oven, the residual gas is trapped by two copper baffles which have holes of 10-mm diameter in the center and are cooled by liquid nitrogen. The liquid nitrogen is stored in two crucible attached to the chamber. The pressure in the vacuum chamber is kept to about $10^{-6}$–$10^{-7}$ Torr.

### 4.2.2 Laser Systems

Two laser systems are essential in the optical double-resonance experiments. In the experiments, the pump laser has to populate the $15650.55$-cm$^{-1}$ level constantly during the measurements. We have the choice of ‘broad-linewidth laser’ or ‘narrow-linewidth laser’ as the pump laser. In former case, the frequency of the pump laser have not to be locked for the first $E_1$ transition, but only to be tuned roughly. In latter case, it have to be locked. Taking into account the smallness of the Stark-induced $E_1$ transition rate, more efficient pumping is preferable. Therefore, we have chosen the latter one.

The wavelength of the pump laser is already fixed to be 638.8 nm, which corresponds to the $E_1$ transition from the $0$-cm$^{-1}$ ($4f^66s^27F_0$) to $15650.55$-cm$^{-1}$ ($4f^66sp^7G_{01}$) levels. A CW ring dye laser with DCM dye is suitable for the wavelength.

For probe laser, we can use whether a dye or a titan-sapphire laser by converting an optics kit in our laboratory. It depends on the wavelength of the probe transition. As the visible-wavelength region of 540–680 nm is tunable with our dye laser, the high-lying odd-parity level $X$ must lie at 30000–34000 cm$^{-1}$. On the other hand, as the infrared-wavelength region is tunable with titan-sapphire laser, the high-lying odd-parity level must lie at 27400–29300 cm$^{-1}$. The criterion of the level $X$ is to decay to the ground or metastable levels rapidly. Since the lifetime of the excited level is proportional to the cube of the transition energy in general, the former slightly has an advantage. However, since the titan-sapphire laser is more stable and powerful than the dye laser, we have decided to use the titan-sapphire laser as the probe laser.

A CW ring dye laser (Spectra-physics Model 380A) is pumped with 6-W power of an argon-ion laser (Spectra-physics Stabilite 2017). The arrangement of the optics in the intracavity is shown in Fig. 4.2. The FM jitter of the laser is about 40 MHz. The output power is about 50 mW at 638.8 nm (DCM) and 200 mW at 573.7 nm (R6G).

Another CW ring laser (Coherent CR899-29) is pumped by 8-W argon-ion laser (Coherent INNOVA-310). The laser has an alternative optics kit of a dye laser or a titanium-sapphire laser. The arrangement of the optics in the intracavity is shown in Fig. 4.3. The frequency of the laser is stabilized within 500 kHz by means of the feedback the error signal provided by an additional reference cavity. The laser is equipped a computer-controlled wavemeter. Then the frequency of the laser can be arbitrarily changed with the precision of 200 MHz through an operation software named as ‘Autoscan II’. The typical output power in single-mode operation is 200–300 mW at 630–675 nm (dye laser with DCM dye) 250–400 mW at 720–825 nm (titanium-sapphire laser). The laser beams are linearly polarized, we can rotate the polarization plane with a half-wave plate ($\lambda/2$).

### 4.2.3 Atomic Beam

The sectional drawing of the atomic beam oven is shown in Fig. 4.4. The crucible is made of molybdenum, and its size is 5 mm$^3 \times$ 50 mm. The orifice at the top of the crucible has 0.6-mm diameter and 5-mm depth. The crucible is attached to one end-plate of the chamber by a tantalum rod of 3-mm diameter and covered with two thermal shields made of tantalum and stainless steel. The insulators are made of boron nitride. The temperature at the oven is monitored with a Pt-Rh thermocouples. The temperature at the indicated position in Fig. 4.4
CHAPTER 4. OVERVIEW OF EXPERIMENTS

Figure 4.2: CW ring dye laser (Spectra-physics Model 380A). Mp: pump mirror, M1, M2, M3, M4: mirrors, BS: beam splitter, PD: photo-diode.

Figure 4.3: CW ring laser (Coherent CR899-29) with (a) dye laser optics kit and (b) tittanium-sapphire laser optics kit. P1, P2, P3: pump mirrors, M1, M2, M4, M5: mirrors, L1: lens, ICA: intracavity etalon assembly.
4.2. APPARATUS

is about 200 K lower than the one inside the crucible. The current flowing in the filament is about 16–18 A. Heating the oven with a tungsten filament up to about 1200 K, the vapor of the neutral atoms was spout out from the orifice. The atomic beam is collimated by several circular slits and went to the interaction points.

4.2.4 Interaction Points

Fluorescence Detection

On the atomic beam line, there exist two points interacting with laser beams. The upstream one is defined as IP1 and the downstream one as IP2, from now on. The locations of IP1 and IP2 from the aperture of the crucible are 395-mm and 595-mm downstream, respectively. IP1 is the main interaction point for the spectroscopy in all experimental stages. IP2 is used only in the optical double-resonance experiments to get the reference signal for the locking system.

The sectional drawing of IP1 is shown in Fig. 4.5. The laser beams are focused and crossed to the atomic beam perpendicularly to reduce the Doppler broadening. The residual Doppler broadening is estimated to be about 6 MHz. The fluorescence from the atoms is collected by a spherical mirror covering the solid angle of $1.38\pi$ sr and detected by a photomultiplier tube (PMT: Hamamatsu R943-02) through an optical filter. The filter is exchangeable, the bandpass and sharp cut filters are prepared. The mirror and the PMT are perpendicularly arranged to both the laser and atomic beams. The PMT is cooled down to $-20^\circ\text{C}$ with the Peltier device and operated in a single photon counting mode.

The sectional drawing of IP2 is shown in Fig. 4.6. The fluorescence from the atoms is collected by an aspherical lens covering the solid angle of $0.25\pi$ sr. The fluorescence collected by this lens is brought out from the chamber and focused on a PMT (Hamamatsu R268) by another lens.
Figure 4.5: Upstream interaction point (IP1).

Figure 4.6: Downstream interaction point (IP2).
4.2. APPARATUS

External Field

At IP1, the external electric field is applied with two optically transparent electrodes made of optical-glass plates, which are coated with ITO (InSnO$_2$). It is possible to apply the high voltage up to 30 kV/cm. One of the electrodes is connected to a DC-voltage power supply (SPELLMAN RHR30PN60/100) for applying the high voltage, and the other is grounded. The direction of the electric field is set perpendicular to both the atomic and the laser beams. The voltage applied to the electrodes is measured with a high-voltage probe and a digital volt meter.

A electrodes unit is composed of a holder, two electrodes and a spherical mirror as shown in Fig. 4.7. We prepare three types of the units; (i) dummy type, (ii) BK7 type and (iii) quartz type. The dummy type has no electrodes and can not generate the field. This type is used in the electric discharge experiments in order to eliminate the effect due to the transmission of the electrodes. The BK7 type has the electrodes made of BK7 plates and AR-coated. This type is transmissive in visible and infrared regions. The quartz type has the electrodes made of quartz plates and no-AR-coated. This type is transmissive in ultraviolet, visible and infrared regions, but is less transmissive than BK7 type in visible and infrared regions. In the electric discharge experiments with dummy type, it is identified which of BK7 and quartz types is effective for detecting the fluorescence. In the $E1$ experiments, BK7 type is used.

The angle between the laser polarization and the applied electric field is adjusted with the half-wave plate to choose a $\pi$ or $\sigma$ transition under the electric field.

The applied electric field at IP1 is calibrated by measuring the Stark splitting of the transition from the 811.92-cm$^{-1}$ to 13999.50-cm$^{-1}$ levels shown in Fig. 4.8. The $\alpha_2$ of 13999.50-cm$^{-1}$ level has been determined precisely to be 38.89(13) kHz/(kV/cm)$^2$ [34]. Although the Stark splitting of 811.92-cm$^{-1}$ level can not be resolved in our experimental system due to the small $\alpha_2$ of 0.27(2) kHz/(kV/cm)$^2$ [44], the uncertainty caused by the Stark splitting of the lower level does not appear by observing the $\pi$ transition. In this situation, two peaks are clearly resolved. Using
CHAPTER 4. OVERVIEW OF EXPERIMENTS

Figure 4.8: Electric-field calibration transition. The electric field is \( \pi \) field.

Eq. 2.36, the splitting energy is given as

\[
\Delta f = \left( -\frac{3}{2} \alpha_{upp}^2 + \frac{1}{4} \alpha_{low}^2 \right) \mathcal{E}^2, \tag{4.1}
\]

where \( \alpha_{upp}^2 \) and \( \alpha_{low}^2 \) are for 13999.50-cm\(^{-1} \) and 811.92-cm\(^{-1} \) level, respectively.

The external magnetic field is applied at IP1 with two Helmholtz-type electromagnets. It generates the magnetic field up to 350 gauss. The direction of the magnetic field is set parallel to the atomic beam and perpendicular to the direction of the electric field. The performance of the magnets is described in ref. [54].

4.2.5 Frequency-calibration System

A fraction of the laser beam is sampled and led to a wavemeter (ADVANTEST TQ8325). The resolution of the measured frequency is 100 MHz. The accuracy is \( \pm 5 \) ppm \( \pm 1 \) count. The first one is the error by measuring in the air. The second one is the quantum error of the electronic counter. Thus the total accuracy is about 100 MHz.

In the \( E_1 \) and electric discharge experiments, iodine molecule (I\(_2\)) vapor cell is used to monitor the atomic-peak position during the measurement. The cell is 20-cm length and 25-mm diameter. The fluorescence or absorption is detected by a PMT (Hamamatsu R268).

The scanning frequency of the laser is monitored with a Fabry-Perot interferometer (FPI). Two types of FPI’s (Coherent Model 216) are prepared. One covers the visible region of 550–650 nm, and the other covers the near infrared region of 690–830 nm. The free spectral range (FSR) of visible-region type has been calibrated to be 297.7(2) MHz by measuring the hyperfine splitting of the \( 5d^26s^4F_{9/2} \) of \( ^{139}\text{La} \) with precisely obtained the hyperfine coupling constants [59]. The FSR of near-infrared region type has been calibrated to be 300.1(3) MHz by measuring the hyperfine splitting of \( ^{85}\text{Rb} \) [60]. The transmitted light of FPI is detected by a PMT (Hamamatsu R2228).

4.2.6 Data-acquisition System

The block diagram of the data acquisition system is shown in Fig. 4.9. The signals from the PMT for the atoms at IP1 are amplified with a fast timing filter amplifier, discriminated and shaped to the logic pulses. The logic pulses are counted with a CAMAC multichannel scaler (MCS), which has 24-bit memory \( \times 32k \) channels.
4.3 Laser Frequency Locking System

4.3.1 Overview

The laser frequency jitter is actively suppressed with a negative feedback to the laser. The negative feedback system consists of three components; the detection of frequency jitter, amplification of frequency control signal and the frequency shifter.

The frequency jitter is detected with a frequency demodulator, which is composed of a frequency reference and a frequency discriminator. The frequency demodulator generates an error signal which is proportional to the difference between the laser frequency and the reference frequency. The error signal is amplified and added to the frequency controller. As a result, the laser frequency is shifted to the frequency reference. The frequency jitter is suppressed with above processes.

Atomic and molecular absorption lines or longitudinal modes of optical cavity are useful for the good frequency reference. In case of the atomic or molecular absorption lines, the linewidth of the spectrum is broadened by the thermal motion of atomic or molecular gas (Doppler broadening). Narrow spectral lines are obtained with several experimental techniques, such as the atomic-beam laser spectroscopy, saturation spectroscopy, cooling of the atoms [61], atomic fountain [62] and ion trap [63]. In case of the optical cavity, although it is not the invariant frequency reference, very narrow linewidth is obtained by using the high-finess cavity.

The dispersion resonance of the laser is usually used as the frequency discriminator. Such
resonance is easily obtained with phase sensitive detection of modulated laser frequency. High sensitivity for detection of resonance information is achieved by optical heterodyne detection with sidebands produced by rf phase modulation, so called frequency-modulation (FM) sideband method [64].

The frequency reference corresponding to 15650.55 cm\(^{-1}\) is necessary in this experiment. We can use whether Sm transition itself or something else, for example absorption of iodine or transmission of FPI. However, since the absorption lines of iodine are sparse around 15650.55 cm\(^{-1}\) [65], it is not rather suitable as the frequency reference of the experiment. Although the broadband FPI has many transmission lines appearing every FSR periodically, it must be highly stabilized to prevent the influences of the thermal change and the vibration. Thus we use the Sm transition as the frequency reference.

The saturation spectroscopy is one of the easiest way for obtaining the narrow frequency reference. However it is difficult to construct the atomic cell of Sm due to rather high boiling point. Special atomic cell of Sm to observe the absorption is reported in Ref. [17]. As the first step of the laser frequency locking system in our setup, we use the fluorescence from atomic beam at IP2 for the frequency reference.

As the fluorescence of Sm is considerably less than those of the alkali atoms, the PMT detecting the fluorescence generates pulses discretely, that is the photon counting mode. The frequency modulation technique, which generates the dispersion curve as the error signal, is adopted with the current mode operation of the PMT in general. By integrating the pulsed signals during a constant time, we can treat the integrated signals like the current mode. However, taking into account the counting rate of the fluorescence of Sm, the integration time should be about 0.1–1 s. The high-speed frequency modulation is not sufficiently reflected in such long integration time. In order to generate the error signals without frequency modulation, we construct the software feedback system using the profile of the fluorescent peak.

### 4.3.2 Principle of Software Feedback

The diagram of the software feedback system is shown in Fig. 4.10. The frequency of the pump laser is controlled with a scanning electronic units (Spectra-physics Model481B), which has two external inputs. The pump beam toward IP2 whose frequency is \(\nu\) interacts with the atomic beam. The output signals from the PMT detecting the fluorescence at IP2 are fed into a ratemeter (EG&G ORTEC Model 449). It integrates the signals during a constant time and have an output proportional to the counting rate. The integration time is set 100 ms to get the enough signal height for locking. The output from the ratemeter is digitized by a 12-bit analog-to-digital converter (ADC: KEITHLEY KPCMCIA-12AI-C) and taken by a personal computer (PC) to control the laser frequency. The ADC has four input dynamic ranges; \(\pm 10 \text{ V}\), \(\pm 5 \text{ V}\), \(\pm 2.5 \text{ V}\) and \(\pm 1.25 \text{ V}\). One of them is chosen depending on the signal height.

The magnitude of the input voltage \((V_{in})\) to the PC is compared with a reference voltage \((V_0)\) corresponding to the middle point of the slope of fluorescent peak for the transition from the 0-cm\(^{-1}\) to 15650.55-cm\(^{-1}\) levels. The compared value just becomes the error signal. The PC generates the laser-frequency control-signal \((V_{out})\) for the laser controller. Previous \(V_{out}\) value is modified by the difference between \((V_{in})\) and \((V_0)\):\n
\[
V_{out} \rightarrow V_{out} + \delta \times (V_{in} - V_0),
\]

where \(\delta\) is gain value determined in the PC.

The laser-frequency control-signals are transmitted from the PC through a 8-bit digital-to-analog converter (DAC: KEITHLEY KPCMCIA-8AOB). The DAC has 8-channel output, and
Figure 4.10: Diagram of the software feedback system. The abbreviations used are as follows. PMT represents a photomultiplier tube, AOD an acousto-optic deflector, SCA: single channel analyzer, PC a personal computer, ADC an analog-to-digital converter, DAC a digital-to-analog converter, Att. an attenuator and Lowpass a lowpass filter. IP1 and IP2 denote upstream and downstream interaction points, respectively.
Figure 4.11: The relation between the frequencies of the peaks at IP1 and IP2; (a) with the first-order diffracted beam, (b) with the zeroth-order diffracted beam
its output dynamic range is ±5 V. One output channel where $V_{out}$ is transmitted is used to control the laser frequency finely through an attenuator of 20 dB and a lowpass filter with the cut frequency of 160 Hz. The another channel is used to control the laser frequency coarsely through an attenuator of 7 dB and a lowpass filter with the cut frequency of 160 Hz. The fine control-signal provides the negative feedback while the coarse control-signal shifts the laser frequency near the fluorescent peak. Each control-signal is fed into the scanning electronics units. As a result, the frequency of the pump laser is locked around the middle point of the slope of the peak ($\nu_{lock}$), the variances of the central frequency of the pump laser are reduced. However, since the time constant of the feedback system is longer than 100 ms, the fast component of the laser-frequency jitter can not be suppressed, and the original linewidth of the pump laser remains.

The pump laser is locked to the peak of $^{152}$Sm whose natural abundance is the largest among seven naturally occurring isotopes. At IP1, the polarization of the pump laser is set perpendicular to the electric field to populate only the sub-levels with $m = \pm 1$.

The frequency of the pump laser toward IP1 is shifted with AOD to follow up the frequency change due to the Stark effect ($\nu + \nu_{AOD}$). In case of the first-order diffracted beam of the AOD, the relation between the frequencies of the peaks at IP1 and IP2 is shown in Fig. 4.11 (a). The frequency shifted by the AOD is given as

$$\nu_{AOD} = \Delta E_{\text{Stark}}(\mathcal{E}, m = \pm 1) - (\nu_{lock} - \nu_0),$$

where $\Delta E_{\text{Stark}}(\mathcal{E}, m = \pm 1)$ is the Stark shift of $m = \pm 1$ components under the electric filed $\mathcal{E}$ (given in Eq. 2.36), and $\nu_0$ corresponds to the resonance frequency without the electric field. The range of the applied electric-field strength is determined from the controllable frequency range of the AOD together with the scalar and tensor polarizabilities of the $15650.55\text{-cm}^{-1}$ level. The electric field of 13–17.5 kV/cm is applied by using the first-order diffracted beam.

The zeroth-order diffracted beam is used for a small electric field. The relation between the frequencies of the peaks at IP1 and IP2 is shown in Fig. 4.11 (b). In case of the zeroth-order diffracted beam, following relation should be satisfied,

$$\Delta E_{\text{Stark}}(\mathcal{E}, m = \pm 1) = \nu_{lock} - \nu_0.$$

---

**Figure 4.12:** Variance of the laser-frequency control signals during the locking; (a) $V_{in}$ and (b) $V_{out}$. 

---

4.3. LASER FREQUENCY LOCKING SYSTEM
CHAPTER 4. OVERVIEW OF EXPERIMENTS

The small electric field near 6 kV/cm can be applied for the zeroth-order diffracted beam.

The variance of $V_{in}$ and $V_{out}$ during the measurements is shown in Fig. 4.12. The $V_0$ is set to 3 V. The $V_{in}$ is localized around the 3 V, and the laser frequency is locked to the $\nu_{lock}$ in Fig. 4.12 (a). At the area (i) in Fig. 4.12 (b), the $V_{out}$ is localized around a certain value, and the locking system is correctly working.

A mode hop of the frequency on the dye laser sometimes appears due to the environmental changes, such as the temperature, electric noise and vibration. As a result, the laser frequency moves to other longitudinal intracavity mode. The FSR of the intracavity is about 200 MHz. The locking system can not follow such discrete laser-frequency change.

When the laser frequency is locked off the peak, the $V_{in}$ corresponding to the tail of the peak generates a negative error signal. This yields the decrease of $V_{out}$, and the laser frequency becomes lower. If the mode hop leads the laser frequency to be higher, it will automatically recover to lock to the peak due to the negative error signal (area (ii) in Fig. 4.12(b)). If the mode hop leads the laser frequency to be lower, the $V_{out}$ goes down to the minimum value of dynamic range. In the operation software, the algorithm is slightly modified to recover from the locking off. If the $V_{out}$ reaches the minimum value, it is changed to the maximum value. And then, the negative error signal decreases the $V_{out}$ until the laser frequency is again locked to the peak (area (iii) in Fig. 4.12 (b)). If the peak exists within the dynamic range of $V_{out}$, the frequency-locking system always recovers.

4.3.3 Performance

It is necessary to calibrate the $V_{out}$ to relative laser frequency in order to investigate the performance of locking system. The transmission spectrum of FPI is measured by changing the $V_{out}$ as shown in Fig. 4.13 (a). The Gaussian function is fitted to all the peaks to determine the central voltages. The linear fit is done for the plots of relative frequencies versus peak-center voltages as shown in Fig. 4.13 (b). The slope of the linear function yields the relation between the $V_{out}$ and the relative laser frequency to be 629.8(79) MHz/V.
4.3. LASER FREQUENCY LOCKING SYSTEM

The $^{152}\text{Sm}$ peak, whose slope plays the role of the frequency discriminator, is shown in Fig. 4.14 (a) by changing the $V_{\text{out}}$. Each point is fitted with a Gaussian function. The variance of $V_{\text{in}}$ during the locking is monitored as shown in Fig. 4.14 (b). The variance of $V_{\text{in}}$ is converted to relative variance of the $V_{\text{out}}$ with the obtained Gaussian function in Fig. 4.14 (a). The variance of the $V_{\text{out}}$ is projected to the histogram as shown in Fig. 4.14 (c). The histogram is fitted with a Gaussian function. The width of the Gaussian function is regarded as the variance of the central frequency of the pump laser. Since the slope of the peak depends on the peak height, the variances of the central frequency are slightly scattered for every experiment. The mean width obtained in different experiment is plotted in Fig. 4.14 (d). As a result, the variance of the central frequency is estimated to be 3.0(7) MHz.

We now consider the locking duration from Fig. 4.14 (b). The laser frequency is locked off the peak only when the mode hop appears. Owing to the algorithm of recovery, it is again locked on the peak every time unless the voltage corresponding to the peak frequency is within the dynamic range of $V_{\text{out}}$. We extract three parameters about the locking duration. The first one is a total locking duration from the beginning until the locking-off due to the out of range of $V_{\text{out}}$. The second one is an individual locking duration from the locking-on until the locking-off due to the mode hop. The last one is a recovery interval from locking-off to locking-on. The mean locking duration of this system is shown in Table 4.1. The first raw represents the mean total locking duration. The second raw represents the mean individual locking duration. The
CHAPTER 4.Overview of Experiments

Table 4.1: Mean locking duration. The first raw represents the mean total locking duration until the locking-off due to the out of range of $V_{out}$. The second raw represents the mean locking duration until the locking-off due to the mode hop. The third raw represents the mean recovery interval from locking-off to locking-on.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Time (sec.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total locking duration</td>
<td>559.7</td>
</tr>
<tr>
<td>Individual locking duration</td>
<td>79.3</td>
</tr>
<tr>
<td>Recovery interval</td>
<td>7.4</td>
</tr>
</tbody>
</table>

third raw represents the mean recovery interval. The total locking duration of our system is about 10 min.

We introduce an Allan variance which is used to evaluate the laser-frequency stability [66]. An observable $y(t)$ is randomly fluctuating as a function of time $t$. We define the time-averaged $y(t)$ as

$$\bar{y}_k \equiv \frac{1}{\tau} \int_{t_k}^{t_k+\tau} y(t) dt \quad (k = 0, 1, 2, \cdots, N),$$

where $\tau$ is a integration time, and $t_{k+1} = t_k + T$. $T$ is a period of the measurement. When $T \simeq \tau$, Allan variance is defined as follows,

$$\sigma_k^2(\tau) \equiv \frac{\langle (\bar{y}_{k+1} - \bar{y}_k)^2 \rangle}{2} = \lim_{N \to \infty} \frac{1}{N} \sum_{k=1}^{N} \frac{(\bar{y}_{k+1} - \bar{y}_k)^2}{2}.$$  \hspace{1cm} (4.6)

The data of the error signal $\delta \times (V_{in} - V_0)$ are converted to the frequencies, and the fluctuation of the frequency is evaluated as the Allan variance. Typical plots of the Allan variance $\sigma$ versus $\tau$ are shown in Fig. 4.15. The $\sigma(\Delta \nu)$ is 3–4 MHz at the largest. This is consistent with the value obtained in Fig. 4.14. It is concluded that the fluctuation of the central frequency of the pump laser is reduced within 3 MHz/10 min.
Chapter 5

E1 Experiments for Low-lying Levels

We perform the spectroscopic study of $4f^66s6p^7G$ term. The purpose of this study is to investigate the spectroscopic properties, such as isotope shift, hyperfine structure, Stark effect and Zeeman effect. The properties of 15650.55-cm$^{-1}$ ($^7G_1$) level, in which our interest lies, are required for the further experimental stage. Moreover, we can expect that the systematic study of $4f^66s6p^7G$ term provides useful information for the PNC measurements.

5.1 Experiments

The overview of experimental setup is shown in Fig. 5.1. The CW ring dye laser (Coherent 899-29) with DCM dye was used for the measurement. The scanning frequency was monitored with the FPI of visible region type. The fluorescence of $I_2$ was measured for monitoring the absolute frequency.

We measured seven $E1$ transitions in this experimental stage. The measured transitions are listed in Table 5.1. Two transitions to the 15650.55-cm$^{-1}$ level were already measured by Kobayashi et al. [56]. We precisely measure these transitions with electric field. We measured the other transitions with no external field, with electric field and with magnetic field.

5.2 Results and Analysis

5.2.1 Relative Frequency Calibration

Typical raw spectra of 638.8-nm transition is shown in Fig. 5.2. The horizontal axis is the MCS channel number. Each vertical axis is MCS counts in every 20 ms. They are (a) the atomic Sm spectrum with simultaneously measured (b) $I_2$ spectrum and (c) FPI transmitted spectrum. The peaks in the $I_2$ spectrum correspond to the rotational-vibrational transitions whose wavenumbers have been precisely determined [65]. They were used as a guide to the positions of Sm peaks during the measurements.

We describe here the procedure of analysis. It is common to the Sm spectra obtained in later experiments. First, all peaks in FPI spectrum are fitted with Gaussian function to determine the central channels of the peaks (Fig. 5.3 (a)), which correspond to the relative frequency of $n \times$ FSR (for nth peak). Second, the plots are made for the relative frequencies versus obtained peak-center channels (Fig. 5.3 (b)). They are fitted with the polynomial functions having the
Figure 5.1: Overview of the experimental setup for $E1$ transition measurements. The abbreviations used are as follows. PMT represents a photomultiplier tube, MCS a multichannel scaler, FPI a Fabry-Perot interferometer and $\lambda/2$ a half-wave plate. IP1 denotes an upstream interaction point.

Table 5.1: Measured $E1$ transition. The abbreviation Config. denotes the configuration. Two transitions to the $4f^66s6p^7G_1$ level were measured with electric field. The other transitions were measured with no external field, with electric field and with magnetic field.

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Lower level</th>
<th>Energy (cm$^{-1}$)</th>
<th>Config. and $J$</th>
<th>Upper level</th>
<th>Energy (cm$^{-1}$)</th>
<th>Config. and $J$</th>
</tr>
</thead>
<tbody>
<tr>
<td>638.8</td>
<td></td>
<td>0.00</td>
<td>$4f^66s^22^7F_0$</td>
<td>15650.55</td>
<td>$4f^66s6p^7G_1$</td>
<td></td>
</tr>
<tr>
<td>650.9</td>
<td></td>
<td>292.58</td>
<td>$4f^66s^22^7F_1$</td>
<td>15650.55</td>
<td>$4f^66s6p^7G_1$</td>
<td></td>
</tr>
<tr>
<td>653.2</td>
<td></td>
<td>811.92</td>
<td>$4f^66s^22^7F_2$</td>
<td>16116.42</td>
<td>$4f^66s6p^7G_2$</td>
<td></td>
</tr>
<tr>
<td>655.2</td>
<td></td>
<td>1489.55</td>
<td>$4f^66s^22^7F_3$</td>
<td>16748.30</td>
<td>$4f^66s6p^7G_3$</td>
<td></td>
</tr>
<tr>
<td>656.4</td>
<td></td>
<td>2273.09</td>
<td>$4f^66s^22^7F_4$</td>
<td>17504.63</td>
<td>$4f^66s6p^7G_4$</td>
<td></td>
</tr>
<tr>
<td>656.6</td>
<td></td>
<td>3125.46</td>
<td>$4f^66s^22^7F_5$</td>
<td>18350.40</td>
<td>$4f^66s6p^7G_5$</td>
<td></td>
</tr>
<tr>
<td>656.3</td>
<td></td>
<td>4020.66</td>
<td>$4f^66s^22^7F_6$</td>
<td>19254.29</td>
<td>$4f^66s6p^7G_6$</td>
<td></td>
</tr>
</tbody>
</table>
5.2. RESULTS AND ANALYSIS

![Typical raw spectra of 638.8-nm transition. The horizontal axis is the MCS channel number. Each vertical axis is MCS counts in every 20 ms; (a) atomic spectrum, (b) spectrum of I_2 rotational-vibrational transition and (c) FPI transmitted spectrum.](image)

order from fourth to eighth. The order of the polynomial minimizing the chi-square is used. The deviations of the data points from the best-fit straight line are plotted in Fig. 5.3 (c). And then, the MCS channels were converted to the relative frequencies. The standard deviation of the data points from the polynomial is regarded as a systematic error of the laser frequency, which is typically 500 kHz. The Sm peaks are fitted with Lorentzian function after subtracting the background. The peak center and FWHM are converted to the relative frequency using the obtained polynomial.

5.2.2 Hyperfine Structure and Isotope Shift

As the even-mass isotopes have the nuclear spin of zero, one peak appears with no external field. The odd-mass isotopes have the hyperfine structure and split into 2J + 1 sub-levels (J ≤ I) or 2I + 1 sub-levels (I ≤ J). Typical spectra of 653.3-nm transition are shown in Fig. 5.4 (a). The spectrum around the odd-mass isotopes of ^{149}Sm and ^{147}Sm is enlarged in Fig. 5.4 (b). The numbers above peaks denote the total angular momenta of lower and upper levels (F_l → F_u) in order.

Using Eq. (2.31), the frequency of a transition between the hyperfine structure levels having
Figure 5.3: Calibration of relative frequency; (a) FPI transmitted peak (b) relative frequency versus FPI peak center and best-fit polynomial (c) deviations of the data points from the best-fit straight line.
5.2. RESULTS AND ANALYSIS

Figure 5.4: Spectra of 653.3-nm transition without external field; (a) the atomic spectrum and (b) the enlargement of atomic spectrum around the odd-mass isotopes of $^{149}\text{Sm}$ and $^{147}\text{Sm}$. The numbers above peaks denote the total angular momenta of lower and upper levels ($F_l \rightarrow F_u$) in order.

the total angular momenta $F_u$ and $F_l$ is expressed as

$$f_{F_uF_l} = \left\{ \frac{1}{2} K(F_u)A_u + L(F_u)B_u \right\} - \left\{ \frac{1}{2} K(F_l)A_l + L(F_l)B_l \right\} + C,$$  \hspace{1cm} (5.1)

where $A_u$, $B_u$, $A_l$ and $B_l$ are the hyperfine coupling constants $A_{hfs}$ and $B_{hfs}$ for upper and lower levels, respectively, and $C$ is the transition frequency. Applying the least-squares method with Eq. (5.1) to the peak-center frequencies of the hyperfine structure multiplets, the parameters $A_u$, $B_u$, $A_l$, $B_l$ and $C$ were determined. About ten spectra were measured for a transition to decrease the statistical uncertainty.

The determined hyperfine coupling constants are listed in Table 5.2. For comparison, the reference values are also listed. They are the values measured by Kobayashi et al. [56] and by Childs and Goodman [43]. The plots of $A_{hfs}$ and $B_{hfs}$ versus $J$ are shown in Fig. 5.5. For $4f^66s6p^7G$ term, the $J$ dependence of $A_{hfs}$ and $B_{hfs}$ constants are found, but $A_{hfs}$ and $B_{hfs}$
Table 5.2: Determined hyperfine coupling constants $A_{\text{hfs}}$ and $B_{\text{hfs}}$ of $^{147}\text{Sm}$ and $^{149}\text{Sm}$ in MHz.

<table>
<thead>
<tr>
<th>Level (cm$^{-1}$)</th>
<th>$^{147}\text{Sm}$</th>
<th>$^{149}\text{Sm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$A_{\text{hfs}}$</td>
<td>$B_{\text{hfs}}$</td>
</tr>
<tr>
<td>292.58</td>
<td>-33.486(45)$^a$</td>
<td>-58.62(43)$^a$</td>
</tr>
<tr>
<td></td>
<td>-33.493(2)$^b$</td>
<td>-58.688(6)$^b$</td>
</tr>
<tr>
<td>811.92</td>
<td>-41.224(81)</td>
<td>-62.24(37)</td>
</tr>
<tr>
<td></td>
<td>-41.186(2)$^b$</td>
<td>-62.229(13)$^b$</td>
</tr>
<tr>
<td>1489.55</td>
<td>-50.235(68)</td>
<td>-33.87(55)</td>
</tr>
<tr>
<td></td>
<td>-50.243(2)$^b$</td>
<td>-33.668(40)$^b$</td>
</tr>
<tr>
<td>2273.09</td>
<td>-59.757(39)</td>
<td>20.30(72)</td>
</tr>
<tr>
<td></td>
<td>-59.707(1)$^b$</td>
<td>21.241(36)$^b$</td>
</tr>
<tr>
<td>3125.46</td>
<td>-69.149(28)</td>
<td>102.83(85)</td>
</tr>
<tr>
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<td>-69.136(1)$^b$</td>
<td>100.608(33)$^b$</td>
</tr>
<tr>
<td>4020.66</td>
<td>-78.494(88)</td>
<td>205.4(16)</td>
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<td>203.432(32)$^b$</td>
</tr>
<tr>
<td>15650.55</td>
<td>-212.63(14)$^c$</td>
<td>-9.58(6)$^a$</td>
</tr>
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<td>16116.42</td>
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<td>-41.91(31)</td>
</tr>
<tr>
<td>16748.30</td>
<td>-82.00(11)</td>
<td>-46.55(33)</td>
</tr>
<tr>
<td>17504.63</td>
<td>-96.475(58)</td>
<td>-29.04(84)</td>
</tr>
<tr>
<td>18350.40</td>
<td>-106.947(41)</td>
<td>27.8(11)</td>
</tr>
<tr>
<td>19254.29</td>
<td>-116.95(12)</td>
<td>103.4(20)</td>
</tr>
</tbody>
</table>

$^a$ Ref. [56]. $^b$ Ref. [43]
5.2. RESULTS AND ANALYSIS

Figure 5.5: $A_{hfs}$ and $B_{hfs}$ constants versus $J$: (a) $4f^66s^2^7F_J$ and (b) $4f^66s6p^7G_J$.

constants do not monotonously change against $J$. This result suggests the configuration mixing on $4f^66s6p^7G$ term.

Using the transition frequency $C$ derived from the analysis of hyperfine structure, we can determine the isotope shift for six isotope pairs. The measured isotope shifts are listed in Table 5.3. Our previous isotope-shift values of $^7F_0 - ^7G_1$ and $^7F_1 - ^7G_1$ transitions [56] are also listed for comparison. We introduce the modified isotope shift $\delta \nu_{\text{MOD}}$: [36,37]

$$\delta \nu_{\text{MOD}} = (\delta \nu_{\text{obs}} - \delta \nu_{\text{NMS}}) \frac{AA'}{A' - A},$$

(5.2)

where $\delta \nu_{\text{obs}}$ is an observed isotope shift. And then the modified isotope shifts of two transitions $i$ and $j$ are connected by the following relation;

$$\delta \nu_{i\text{MOD}} = \frac{E_i}{E_j} \delta \nu_{j\text{MOD}} + \left( M_{i\text{SMS}} - \frac{E_i}{E_j} M_{j\text{SMS}} \right).$$

(5.3)

The two-dimensional plot of the modified isotope shifts for the transition $i$ and $j$ is called as King plot. Its slope gives the ratio $E_i/E_j$ as shown in Eq. (5.3), and the intercept of the line
**Table 5.3:** Measured isotope shift in MHz between the two isotopes $A - A'$.

<table>
<thead>
<tr>
<th>Transition (nm)</th>
<th>154-152</th>
<th>152-150</th>
<th>150-148</th>
<th>148-144</th>
<th>149-148</th>
<th>148-147</th>
</tr>
</thead>
<tbody>
<tr>
<td>638.8</td>
<td>-1250.68(7)</td>
<td>-2320.74(7)</td>
<td>-1651.91(7)</td>
<td>-2813.35(8)</td>
<td>-498.16(6)</td>
<td>-830.22(6)</td>
</tr>
<tr>
<td>650.9</td>
<td>-1253.95(56)</td>
<td>-2328.03(56)</td>
<td>-1655.72(56)</td>
<td>-2822.40(56)</td>
<td>-498.84(43)</td>
<td>-833.50(45)</td>
</tr>
<tr>
<td>653.2</td>
<td>-1257.23(17)</td>
<td>-2335.97(17)</td>
<td>-1661.39(17)</td>
<td>-2829.64(17)</td>
<td>-499.62(13)</td>
<td>-836.02(13)</td>
</tr>
<tr>
<td>655.2</td>
<td>-1254.44(27)</td>
<td>-2329.70(27)</td>
<td>-1657.70(27)</td>
<td>-2824.53(27)</td>
<td>-499.94(20)</td>
<td>-833.33(20)</td>
</tr>
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<td>656.4</td>
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<td>-2329.67(20)</td>
<td>-1657.67(20)</td>
<td>-2823.73(20)</td>
<td>-499.51(15)</td>
<td>-833.58(15)</td>
</tr>
<tr>
<td>656.6</td>
<td>-1254.96(39)</td>
<td>-2331.59(39)</td>
<td>-1657.55(39)</td>
<td>-2824.03(39)</td>
<td>-498.88(28)</td>
<td>-833.86(28)</td>
</tr>
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<td>656.3</td>
<td>-1256.90(22)</td>
<td>-2336.38(22)</td>
<td>-1661.91(22)</td>
<td>-2827.90(22)</td>
<td>-501.36(16)</td>
<td>-833.65(17)</td>
</tr>
</tbody>
</table>

a) Ref. [56].

---

**Figure 5.6:** King plot for 653.2-nm transition. The 591.64-nm ($^7F_1-^7F_2$) transition in ref. [37] is taken as a reference for the horizontal axis of King plot. Each value of modified isotope shift is normalized to that of 154-152 isotope pair.

The modified isotope shift, $\Delta \omega$, for a measured transition is given by $\Delta \omega = M_{SMS} - (E_i/E_j)M_{SMS}$. The linearity of this plot is also useful to examine the reliability of the experimental values of isotope shift. The King plot for 653.2-nm transition is shown in Fig. 5.6. The 591.64-nm ($^7F_1-^7F_2$) transition in ref. [37] is taken as a reference for the horizontal axis of King plot. Each value of modified isotope shift is normalized to that of 154-152 isotope pair. Since both the reference transition and the measured transitions belong to $ns^2-nsnp$ transition, the King plot was best-fitted with a straight line passing through the origin of the coordinates. Using the value $E_j = -0.285(11)$ and $f(Z) = 19.3$ GHz/fm$^2$ in Ref. [37], we can obtain SMS and FS as shown in Table 5.4. These values yield $\Delta |\psi(0)|_i^2$ and nuclear parameter $\lambda$ for each transition. They are listed in Table 5.5 and Table 5.6.

**5.2.3 Stark Splitting**

We could observe the Stark splittings for 638.8-nm, 650.9-nm, 653.2-nm and 655.2-nm transitions. Stark splittings for other transitions were not resolved in our experimental resolution.
5.2. RESULTS AND ANALYSIS

Table 5.4: NMS, SMS and FS between the two isotopes A - A’. All values are given in MHz.

<table>
<thead>
<tr>
<th>Transition (nm)</th>
<th>Isotope pair</th>
<th>154-152</th>
<th>152-150</th>
<th>150-148</th>
<th>148-144</th>
<th>149-148</th>
<th>148-147</th>
</tr>
</thead>
<tbody>
<tr>
<td>638.8 a)</td>
<td>NMS</td>
<td>21.8</td>
<td>22.4</td>
<td>23.0</td>
<td>48.0</td>
<td>11.6</td>
<td>11.7</td>
</tr>
<tr>
<td></td>
<td>SMS</td>
<td>0(11)</td>
<td>0(11)</td>
<td>0(12)</td>
<td>0(24)</td>
<td>0.0(58)</td>
<td>0.0(59)</td>
</tr>
<tr>
<td></td>
<td>FS</td>
<td>-1273(11)</td>
<td>-2343(11)</td>
<td>-1675(12)</td>
<td>-2861(24)</td>
<td>-509.7(58)</td>
<td>-842.0(59)</td>
</tr>
<tr>
<td>650.9 a)</td>
<td>NMS</td>
<td>21.4</td>
<td>22.0</td>
<td>22.6</td>
<td>47.1</td>
<td>11.4</td>
<td>11.5</td>
</tr>
<tr>
<td></td>
<td>SMS</td>
<td>0(11)</td>
<td>0(11)</td>
<td>0(11)</td>
<td>0(24)</td>
<td>0.0(57)</td>
<td>0.0(58)</td>
</tr>
<tr>
<td></td>
<td>FS</td>
<td>-1275(11)</td>
<td>-2350(11)</td>
<td>-1678(11)</td>
<td>-2869(24)</td>
<td>-510.2(57)</td>
<td>-845.0(58)</td>
</tr>
<tr>
<td>653.2</td>
<td>NMS</td>
<td>21.4</td>
<td>21.9</td>
<td>22.5</td>
<td>46.9</td>
<td>11.3</td>
<td>11.5</td>
</tr>
<tr>
<td></td>
<td>SMS</td>
<td>0(11)</td>
<td>0(11)</td>
<td>0(11)</td>
<td>0(23)</td>
<td>0.0(57)</td>
<td>0.0(57)</td>
</tr>
<tr>
<td></td>
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<td>-2358(11)</td>
<td>-1684(11)</td>
<td>-2877(24)</td>
<td>-511.0(57)</td>
<td>-847.5(58)</td>
</tr>
<tr>
<td>655.2</td>
<td>NMS</td>
<td>21.3</td>
<td>21.9</td>
<td>22.4</td>
<td>46.8</td>
<td>11.3</td>
<td>11.5</td>
</tr>
<tr>
<td></td>
<td>SMS</td>
<td>0(11)</td>
<td>0(11)</td>
<td>0(11)</td>
<td>0(23)</td>
<td>0.0(57)</td>
<td>0.0(57)</td>
</tr>
<tr>
<td></td>
<td>FS</td>
<td>-1276(11)</td>
<td>-2352(11)</td>
<td>-1680(11)</td>
<td>-2871(23)</td>
<td>-511.2(57)</td>
<td>-844.8(58)</td>
</tr>
<tr>
<td>656.4</td>
<td>NMS</td>
<td>21.2</td>
<td>21.8</td>
<td>22.4</td>
<td>46.7</td>
<td>11.3</td>
<td>11.4</td>
</tr>
<tr>
<td></td>
<td>SMS</td>
<td>0(11)</td>
<td>0(11)</td>
<td>0(11)</td>
<td>0(23)</td>
<td>0.0(56)</td>
<td>0.0(57)</td>
</tr>
<tr>
<td></td>
<td>FS</td>
<td>-1276(11)</td>
<td>-2351(11)</td>
<td>-1680(11)</td>
<td>-2870(23)</td>
<td>-510.8(57)</td>
<td>-845.0(57)</td>
</tr>
<tr>
<td>656.6</td>
<td>NMS</td>
<td>21.3</td>
<td>21.8</td>
<td>22.4</td>
<td>46.7</td>
<td>11.3</td>
<td>11.4</td>
</tr>
<tr>
<td></td>
<td>SMS</td>
<td>0(11)</td>
<td>0(11)</td>
<td>0(12)</td>
<td>0(23)</td>
<td>0.0(56)</td>
<td>0.0(57)</td>
</tr>
<tr>
<td></td>
<td>FS</td>
<td>-1276(11)</td>
<td>-2353(11)</td>
<td>-1680(11)</td>
<td>-2871(23)</td>
<td>-510.1(57)</td>
<td>-845.3(57)</td>
</tr>
<tr>
<td>656.3</td>
<td>NMS</td>
<td>21.3</td>
<td>21.8</td>
<td>22.4</td>
<td>46.7</td>
<td>11.3</td>
<td>11.4</td>
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<tr>
<td></td>
<td>SMS</td>
<td>0(11)</td>
<td>0(11)</td>
<td>0(11)</td>
<td>0(23)</td>
<td>0.0(56)</td>
<td>0.0(57)</td>
</tr>
<tr>
<td></td>
<td>FS</td>
<td>-1278(11)</td>
<td>-2358(11)</td>
<td>-1684(11)</td>
<td>-2875(23)</td>
<td>-512.6(56)</td>
<td>-845.1(57)</td>
</tr>
</tbody>
</table>

a) Ref. [56].

Table 5.5: Obtained $\Delta|\psi(0)|^2$ values.

| Transition (nm) | $\Delta|\psi(0)|^2$ $(\times 10^{-14}/$fm$^3)$ |
|-----------------|---------------------------------------------|
| 638.8           | -3.81(15)                                   |
| 650.9           | -3.82(15)                                   |
| 653.2           | -3.83(15)                                   |
| 655.2           | -3.81(15)                                   |
| 656.4           | -3.81(15)                                   |
| 656.6           | -3.81(15)                                   |
| 656.3           | -3.82(15)                                   |
Table 5.6: Obtained nuclear parameter $\lambda$ in fm$^2$.

<table>
<thead>
<tr>
<th>Transition (nm)</th>
<th>Isotope pair</th>
<th>154-152</th>
<th>152-150</th>
<th>150-148</th>
<th>148-144</th>
<th>149-148</th>
<th>148-147</th>
</tr>
</thead>
<tbody>
<tr>
<td>638.8</td>
<td>-0.2301(34) -0.4237(54) -0.3028(41) -0.5174(75) -0.0922(15) -0.1522(21)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>650.9</td>
<td>-0.2299(33) -0.4236(54) -0.3026(41) -0.5173(74) -0.0920(15) -0.1523(21)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>653.2</td>
<td>-0.2305(33) -0.4251(54) -0.3036(41) -0.5186(74) -0.0921(15) -0.1528(21)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>655.2</td>
<td>-0.2305(33) -0.4249(54) -0.3036(41) -0.5188(74) -0.0924(15) -0.1527(21)</td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>656.4</td>
<td>-0.2306(33) -0.4249(54) -0.3036(41) -0.5187(74) -0.0923(15) -0.1527(21)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>656.6</td>
<td>-0.2305(33) -0.4251(54) -0.3035(41) -0.5184(74) -0.0922(15) -0.1524(21)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>656.3</td>
<td>-0.2306(33) -0.4252(54) -0.3037(41) -0.5182(74) -0.0923(15) -0.1524(21)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The typical spectra of 638.8-nm transition with electric field are shown in Fig. 5.7. The Stark splitting of $^{152}\text{Sm}$ peak is shown in Fig. 5.7 (b) and (c). The $\lambda/2$ plate was set at 157.5° for all transitions. Since the 638.8-nm transition has the lower level of electronic total angular momentum $J = 0$, the lower level does not split. For the other transitions in which the Stark splittings were observed, the splittings of lower levels were not resolved because of small $\alpha_2$ [44] in our experimental resolution. The energy shift of a fine structure level is given in Eq. (2.36). Assuming that the splitting energy of lower level is negligibly small, splitting energy between the peaks with magnetic quantum numbers $m$ and $m'$ in upper level is written as

$$\Delta E_{m,m'} = -\frac{3(m^2 - m'^2)}{2J(2J - 1)}\alpha_2 E^2.$$  (5.4)

About twenty spectra were taken for a transition. The electric field was applied up to 28 kV/cm.

Table 5.7: Obtained $\alpha_2$ in kHz/(kV/cm)$^2$. The values in square bracket are the errors taking the uncertainty of the gap into account.

<table>
<thead>
<tr>
<th></th>
<th>15650.55 cm$^{-1}$</th>
<th>1611.042 cm$^{-1}$</th>
<th>1674.830 cm$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>638.8-nm</td>
<td>650.9-nm</td>
<td>Average</td>
</tr>
<tr>
<td>$^{154}\text{Sm}$</td>
<td>-546.40(24)</td>
<td>-546.39(45)</td>
<td>-546.40(21)</td>
</tr>
<tr>
<td>$^{152}\text{Sm}$</td>
<td>-546.50(24)</td>
<td>-546.54(45)</td>
<td>-546.51(21)</td>
</tr>
<tr>
<td>$^{150}\text{Sm}$</td>
<td>-547.53(25)</td>
<td>-548.03(45)</td>
<td>-547.65(21)</td>
</tr>
<tr>
<td>$^{148}\text{Sm}$</td>
<td>-548.14(24)</td>
<td>-549.16(45)</td>
<td>-548.38(21)</td>
</tr>
<tr>
<td>$^{144}\text{Sm}$</td>
<td>-549.46(25)</td>
<td>-549.97(45)</td>
<td>-549.58(21)</td>
</tr>
</tbody>
</table>

As shown in Eq. (5.4), Stark-splitting energy is proportional to the square of electric field strength $E^2$. The plots of splitting energy versus $E^2$ are shown in Fig. 5.8. This is the result of $^{152}\text{Sm}$ for 638.8-nm transition. The best-fit was obtained with a straight line passing through the origin of the coordinates, and the $\alpha_2$ was derived from the slope for each isotope. The obtained $\alpha_2$ values are listed in Table 5.7, and the plots of $\alpha_2$ versus mass number are shown in Fig. 5.9. For 15650.55-cm$^{-1}$ level, the $\alpha_2$ values are obtained as the weighted average of the two values derived from 638.8-nm and 650.9-nm transitions. In Table 5.7, the values in square
5.2. RESULTS AND ANALYSIS

Figure 5.7: Typical spectra of 638.8-nm transition with electric field: (a) Stark splitting of all isotope peaks, (b) the peak of $^{152}\text{Sm}$ with no external field and (c) with electric field. The $\lambda/2$ plate was set at $157.5^\circ$.

Figure 5.8: Dependence of the Stark splitting of the $^{152}\text{Sm}$ peak in the 638.8-nm transition on the square of electric field strength.
Figure 5.9: Dependence of $\alpha_2$ values on mass number for (a) 15650.55-cm$^{-1}$ level, (b) 1611.042-cm$^{-1}$ level and (c) 16748.30-cm$^{-1}$ level.
5.2. RESULTS AND ANALYSIS

Table 5.8: Obtained g-values. The values in square bracket are the uncertainty taking the errors of $g_l$ into account.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$4f^66s6p,^7G_2$</th>
<th>$4f^66s6p,^7G_3$</th>
<th>$4f^66s6p,^7G_4$</th>
<th>$4f^66s6p,^7G_5$</th>
<th>$4f^66s6p,^7G_6$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{154}\text{Sm}$</td>
<td>0.96474(20)</td>
<td>1.27177(10)</td>
<td>1.399997(27)</td>
<td>1.447343(46)</td>
<td>1.466064(72)</td>
</tr>
<tr>
<td>$^{152}\text{Sm}$</td>
<td>0.96479(20)</td>
<td>1.27184(10)</td>
<td>1.399945(27)</td>
<td>1.447344(47)</td>
<td>1.466131(38)</td>
</tr>
<tr>
<td>$^{150}\text{Sm}$</td>
<td>0.96476(20)</td>
<td>1.271800(77)</td>
<td>1.399971(34)</td>
<td>1.447344(51)</td>
<td>1.466115(52)</td>
</tr>
<tr>
<td>$^{148}\text{Sm}$</td>
<td>0.96476(20)</td>
<td>1.271804(77)</td>
<td>1.399971(34)</td>
<td>1.447344(51)</td>
<td>1.466115(52)</td>
</tr>
<tr>
<td>$^{144}\text{Sm}$</td>
<td>0.96491(20)</td>
<td>1.271804(77)</td>
<td>1.399971(34)</td>
<td>1.447344(51)</td>
<td>1.466115(52)</td>
</tr>
<tr>
<td>Average</td>
<td>0.96479(90)</td>
<td>1.271804(77)</td>
<td>1.399971(34)</td>
<td>1.447344(51)</td>
<td>1.466115(52)</td>
</tr>
<tr>
<td>Ref. [42]</td>
<td>0.960</td>
<td>1.27</td>
<td>1.395</td>
<td>1.44</td>
<td>1.47</td>
</tr>
</tbody>
</table>

bracket are the errors taking the uncertainty of electrode gap into account. We can clearly find the isotope dependence of $\alpha_2$ for 15650.55-cm$^{-1}$ level in Fig. 5.9 (a).

5.2.4 Zeeman Splitting

Zeeman splittings were resolved for all transitions. The typical spectra of 653.2-nm transition with magnetic field are shown in Fig. 5.10 (a). The Zeeman splitting of $^{152}\text{Sm}$ peak is shown in Fig. 5.10 (b) and (c). The numbers above each peak with magnetic field denote the magnetic quantum numbers of lower and upper levels ($m_l \rightarrow m_u$) in order. Since the peaks of $\pi$ components ($\Delta m = 0$) overlap with those of $\sigma$ components ($\Delta m = \pm 1$) in general setup of the directions between laser polarization and magnetic field, the $\lambda/2$ plate was set at 135$^\circ$ to simplify the analysis for 653.2-nm transition. With this setting, the direction of laser polarization was set parallel to that of the magnetic field, and $\pi$ components should be vanished. However, they weakly appeared in the figure. This is possibly caused by the inhomogeneous magnetic filed or small misalignment of the laser polarization. For the other transitions, the $\lambda/2$ plate was set at 157.5$^\circ$, both the $\pi$ components and $\sigma$ components appeared.

In the weak field limit, the energy shift of a fine structure level is given in Eq. (2.43). Then the transition frequency between Zeeman sub-levels with magnetic field strength $B$ can be written as

$$f_{m_u,m_l} = \mu_B(g_u m_u - g_l m_l)B + C,$$

where $g_u$ and $m_u$ are the g-value and the magnetic quantum number for the upper level, respectively, $g_l$ and $m_l$ are those for the lower level, and $C$ is the original frequency of the transition without external field. About twenty spectra were taken for a transition. The magnetic field was applied up to 350 gauss.

The g-values for the $4f^66s^2\,^7F$ ground terms were accurately measured by the atomic-beam magnetic-resonance method [43]. Therefore the $g_l$ and $\mu_B$ were fixed, and the least-squares method was applied using Eq. (5.5) with free parameters $g_u$, $B$ and $C$. With this analysis, we could determine not only the $g_u$ values, but also the magnetic field strength at the interaction point precisely.

The obtained g-values for isotopes are listed in Table 5.8. The blank fields represent that the intensity of each peak was too weak to analyze, or the peaks were buried in hyperfine peaks. The errors of averaged g-values are the uncertainty taking the errors of lower level’s g-values into account. The g-values in ref. [42] are also listed. They are in good agreement with the present values.
Figure 5.10: Typical spectra of 653.2-nm transition with magnetic field; (a) Zeeman splitting of all isotope peaks, (b) the peak of $^{152}$Sm with no external field and (c) with magnetic field. The numbers above each peak with magnetic field denote the magnetic quantum numbers of lower and upper levels ($m_l \rightarrow m_u$) in order. The $\lambda/2$ plate was set at $135^\circ$, that is, the direction of laser polarization was parallel to that of magnetic field. In this situation, $\pi$ components ($\Delta m = 0$) must be vanished, but weakly appeared.
Figure 5.11: $g$-value versus magnetic field strength for $^{152}\text{Sm}$ in 653.2-nm transition.

The plots of $g$-value versus magnetic field strength for $^{152}\text{Sm}$ in 653.2-nm transition are shown in Fig. 5.11. We can not find the magnetic field dependence of $g$-value does not appear. Thus the Paschen-Back effect is negligible for the magnetic field strength applied in this experiment.
Chapter 6

Electric Discharge Experiments for High-lying Levels

The purpose of this experimental stage is to explore the high-lying odd-parity levels which are suitable for the optical double-resonance experiments. Since the transition probability is proportional to the cube of the transition energy in general, the transition with larger energy has shorter lifetime. Therefore the requirement of the suitable levels is that they decay to the ground or metastable levels rapidly.

In order to investigate the decay process of the levels, the electric discharge method is appropriate. In the technique, after the atoms are populated on a metastable level by colliding the thermal electrons, the atoms are excited to high-lying odd-parity levels with one laser. The description of the technique is given in Ref. [48]. The fluorescence from the level is detected through a bandpass filter (Fig. 6.1). The wavelength of the fluorescence is identified changing the wavelength window of the filter.

6.1 Experiments

The overview of the experimental setup based on electric discharge is shown in Fig. 6.2. The atomic beam oven is slightly modified from the normal type to generate the atomic beam populated on the metastable level (10801.10 cm\(^{-1}\) \(4f^{6}5d6s\, ^{9}H_{1}\)). The atomic beam oven furnished with a cathode for electric discharge is shown in Fig. 6.3. The cathode is made of tantalum wire with 0.5-mm diameter and has the form of 8-turn helical coil of 5-mm diameter. The crucible itself plays a role of the discharge anode. The electrons are discharged from the cathode toward the top of the crucible and collide with atoms. This collision yields the atomic beam populated on metastable levels. \(V_C\) and \(V_D\) are cathode and discharge voltages, respectively. The power supply for \(V_C\) is operated with constant current mode, and the typical current is 5 A. The power supply for \(V_D\) is operated with constant voltage mode, and the typical voltage is 10–30 V.

Five bandpass and one sharp-cut filters are prepared for the experiments. Each bandpass filter has the bandwidth of 100-nm. The central wavelengths of each filter are 350 nm, 450 nm, 550 nm, 650 nm and 750 nm, respectively (denoted as BPF350, BPF450, BPF550, BPF650 and BPF750). The sharp-cut filter blocks the light whose wavelength is shorter than 800 nm (denoted as SCF800).

Since the titan-sapphire laser is used as the probe laser in the optical double-resonance
6.1. EXPERIMENTS

Figure 6.1: Principle of the electric discharge experiment. The atoms are populated on a metastable level by colliding the thermal electrons and are excited to high-lying odd-parity levels with one laser. The fluorescence from the level is detected through a bandpass filter.

Figure 6.2: Overview of the experimental setup based on electric discharge. The abbreviations used are as follows. PMT represents a photomultiplier tube, MCS a multichannel scaler and FPI a Fabry-Perot interferometer. IP1 denotes an upstream interaction point.
experiments, the level $X$ lies at $27400$–$29300$ cm$^{-1}$. To populate the level from the $10801.10$-cm$^{-1}$ level, the laser with the wavelength of $540$–$600$ nm is required. Rhodamine 110 (R110) and Rhodamine 6G (R6G) dyes cover the wavelength of $540$–$575$ nm and $570$–$620$ nm, respectively. In order to choose the suitable level $X$, we put additional restrictions among the levels in the region of $27400$–$29300$ cm$^{-1}$;

- the probe laser having the wavelength around central wavelength of the titan-sapphire laser (780 nm), and

- the level with electronic angular momentum $J$ of 0 or 1.

The first one is required for getting the high-power and stable beam. The second one is to avoid the reduction of the peak counting rate caused by the complex fine structure splitting. Under above restrictions, we choose the candidate for the high-lying levels as listed in Table 6.1. The third column is the wavelength of the $E1$ transition from $10801.10$-cm$^{-1}$ level in the electric discharge experiments. The fourth column is the wavelength of the Stark-induced $E1$ transition from $15650.55$-cm$^{-1}$ level in the optical double-resonance experiments.

The fluorescence from the high-lying odd-parity levels is studied as follows. The wavelength of the decay transitions to the ground levels are within $300$–$400$ nm, where is covered with the BPF350. In case of the BPF450, the decay transitions to the levels around $3000$–$8000$ cm$^{-1}$ corresponds to the wavelength in $400$–$500$ nm. However, there are not such levels with $J=0$–$2$ (from the restriction of $E1$ selection rule). The BPF550 covers the decay transitions to the metastable levels $(4f^6d6s^9H_{1,2})$. In case of the BPF650, BPF750 and SCF800, there are many possible decay transitions having the wavelength longer than $600$ nm.

In addition to the decay process, another information may be obtained in this experiment. One of our interests is to search a high-lying odd-parity level $X$ to which the $E1$ transition amplitude from the $15639.80$-cm$^{-1}$ level is large. Taking into account the leading configuration, the $E1$ transition amplitudes from the $15639.80$-cm$^{-1}$ to high-lying levels listed in Table 6.1 are the type of $4f^6d6s$–$4f^5d6s^2$, which is forbidden. In this situation, we have to expect the configuration mixing on the levels. The leading-configuration percentages of high-lying odd-parity levels are not investigated sufficiently while that of $15639.80$-cm$^{-1}$ level is well-known to be $58\%$ [42].

The $10801.10$-cm$^{-1}$ metastable level has the same configuration of the $15639.80$-cm$^{-1}$ level. Thus the transition from the $10801.10$-cm$^{-1}$ to listed high-lying levels are forbidden too. As the leading-configuration percentage of $10801.10$-cm$^{-1}$ level is well-known to be $99\%$ [42], the
6.2. RESULTS AND ANALYSIS

Table 6.1: Candidate for the high-lying levels in optical double-resonance experiments. The wavelength of the $E1$ transition from 10801.10-cm$^{-1}$ level with electric discharge is shown in the third column. The wavelength of the Stark-induced $E1$ transition from 15650.55-cm$^{-1}$ level with optical double-resonance is shown in the fourth column.

<table>
<thead>
<tr>
<th>High-lying odd-parity level</th>
<th>$E1$ with ED</th>
<th>$E1_{Stark}$ with ODR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy (cm$^{-1}$)</td>
<td>Config. and J</td>
<td>Wavelength (nm)</td>
</tr>
<tr>
<td>27888.93</td>
<td>$4f^55d6s^2,^7D_1$</td>
<td>585.2</td>
</tr>
<tr>
<td>27925.31</td>
<td>$4f^55d6s^2,J=0$</td>
<td>584.0</td>
</tr>
<tr>
<td>28168.22</td>
<td>$4f^55d6s^2,J=0$</td>
<td>575.8</td>
</tr>
<tr>
<td>28233.08</td>
<td>$4f^55d6s^2,J=1$</td>
<td>573.7</td>
</tr>
</tbody>
</table>

observed transition in the discharge experiments is owing to the configuration mixing on the high-lying levels. The strength of these transitions may give us the priority for searching the large Stark-induced $E1$ amplitudes.

6.2 Results and Analysis

The fluorescence was observed for the 575.8-nm and 573.7-nm transitions among the transitions listed in Table 6.1. Although the fluorescence was observed for the 584.0-nm transition, observed peaks were quite small. As a result, two levels of 28168.22 cm$^{-1}$ and 28233.08 cm$^{-1}$ were chosen as the candidates for the optical double-resonance experiment. In addition to these levels, we have decided to measure the 28704.25-cm$^{-1}$ level ($4f^55d6s^2\,J=1$). The Stark-induced $E1$ transition to the level has the wavelength of 765.9 nm, which does not satisfy the restriction to be around central wavelength of the titan-sapphire laser (780 nm). We use this level to check the contribution of low-lying level to the Stark-induced $E1$ transition. Kuwamoto has already performed the Stark spectroscopy for this level with the electric discharge method [47, 48]. It found that $\alpha_2$ of the level was too small to resolve the Stark splitting in our experimental system in spite of existence of close-lying neighbor. Thus this high-lying level does not contribute to the Stark-induced $E1$ transition strongly.

Typical spectrum of 573.7-nm transition with several wavelength windows are shown in Fig. 6.4. They are the spectrum with five kinds of bandpass windows ((a) 300–400-nm, (b) 400–500-nm, (c) 500–600-nm, (d) 600–700-nm, (e) 700–800-nm) and (f) window longer than the wavelength of 800 nm. The fluorescence was observed with 300–400-nm and 500–600-nm bandpass windows. The filters enable us to identify the level energy to which atoms decay. The level energy covered with the wavelength windows are listed in Table 6.2. The decay transitions to the ground and metastable levels are covered with 300–400-nm and 500–600-nm windows, respectively.

We estimate the decay branching ratio of the high-lying odd-parity levels for each optical filter. In this estimation, we take into account the transmissions of quartz window and optical filters, reflectance of mirror and quantum efficiency of PMT for each wavelength. The upper limits for the wavelengths where the fluorescence was not observed are determined from the fluctuation of background level. This result suggests that transitions having the wavelength of 500–600 nm account for the most part of the decay processes of these levels. Thus we choose the BK7 type electrodes unit, which is efficient in visible wavelength region, for the optical
Figure 6.4: Spectrum of 573.7-nm transition with several wavelength windows; (a) with 300–400-nm window, (b) with 400–500-nm window, (c) with 500–600-nm window, (d) with 600–700-nm window, (e) with 700–800-nm window and (f) with window longer than 800 nm. The fluorescence was observed in (a) and (c).
6.2. RESULTS AND ANALYSIS

Table 6.2: Energy range covered with bandpass windows from the high-lying odd-parity level.

<table>
<thead>
<tr>
<th>Window</th>
<th>from 28168.22 cm(^{-1})</th>
<th>from 28233.08 cm(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>300–400 nm</td>
<td>0.00–3168.22 cm(^{-1})</td>
<td>0.00–3233.08 cm(^{-1})</td>
</tr>
<tr>
<td>400–500 nm</td>
<td>3168.22–8168.22 cm(^{-1})</td>
<td>3233.08–8233.08 cm(^{-1})</td>
</tr>
<tr>
<td>500–600 nm</td>
<td>8168.22–11501.55 cm(^{-1})</td>
<td>8233.08–11566.41 cm(^{-1})</td>
</tr>
<tr>
<td>600–700 nm</td>
<td>11501.55–13882.51 cm(^{-1})</td>
<td>11566.41–13947.37 cm(^{-1})</td>
</tr>
<tr>
<td>700–800 nm</td>
<td>13882.51–15668.22 cm(^{-1})</td>
<td>13947.37–15733.08 cm(^{-1})</td>
</tr>
<tr>
<td>≥ 800 nm</td>
<td>15668.22–28168.22 cm(^{-1})</td>
<td>15733.08–28233.08 cm(^{-1})</td>
</tr>
</tbody>
</table>

Table 6.3: Decay branching ratio of high-lying odd-parity levels.

<table>
<thead>
<tr>
<th>Wavelength</th>
<th>28168.22 cm(^{-1})</th>
<th>28233.08 cm(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>300–400 nm</td>
<td>0.0027(20)</td>
<td>0.010(21)</td>
</tr>
<tr>
<td>400–500 nm</td>
<td>≤ 0.0059</td>
<td>≤ 0.0038</td>
</tr>
<tr>
<td>500–600 nm</td>
<td>0.997(+1(-57))</td>
<td>0.983(+13(-34))</td>
</tr>
<tr>
<td>600–700 nm</td>
<td>≤ 0.0065</td>
<td>≤ 0.0041</td>
</tr>
<tr>
<td>700–800 nm</td>
<td>≤ 0.0040</td>
<td>≤ 0.0019</td>
</tr>
<tr>
<td>≥ 800 nm</td>
<td>≤ 0.0025</td>
<td>≤ 0.0053</td>
</tr>
</tbody>
</table>

do double-resonance experiments.

As the transition probability is generally proportional to the cube of the transition energy, it is anomalous that the branching ratio of the 500–600-nm transitions is much larger than that of the 300–400-nm transitions. Taking into account the leading configurations, the \(E1\) transition amplitudes from the high-lying levels to ground and metastable levels are the type of \(4f^55d6s^2\)–\(4f^66s^2\) and \(4f^55d6s^2\)–\(4f^65d6s\), respectively. In general, the former is allowed while the latter is forbidden. However, if the high-lying levels rather include the configurations of \(4f^65d6p\) or \(4f^66s6p\), the \(E1\) transition amplitudes to the metastable levels would be allowed and considerably larger than those to the ground levels.
Chapter 7

Optical Double-resonance Experiments

7.1 Experiments

The overview of experimental setup of the optical double resonance is shown in Fig. 7.1. The central frequency of the pump laser is monitored with the wavemeter. The beam of the pump laser is divided into two paths. One is led to IP2 to get a reference signal to lock the frequency of the pump laser to $E_1$ transition from the 0-cm$^{-1}$ to 15650.55-cm$^{-1}$ levels. The other is led to IP1 to populate the 15650.55-cm$^{-1}$ level. The pump beam toward IP1 passes through an acousto-optic deflector (AOD: HOYA SCHOT AOD-200). In order to modify the laser pass diffracted by the AOD, the pump beam passes through the AOD two times contrarily via a polarized beam splitter (PBS), a quarter-wave plate ($\lambda/4$), a convex lens and a mirror. The frequency of the first-order diffracted beam is shifted from 120 to 200 MHz.

Assuming the atomic beam velocity to be 400 m/s, the atomic beam travels about 1 mm during the lifetime of 15650.55-cm$^{-1}$ level of 2.626(17) $\mu$m [34]. Thus the pump and probe beams are merged to the completely same path with a dichroic mirror (DM). It is reflective for visible light and transmissive for the infrared light. The reflectance of the DM is 99.2% at 640 nm. The transmission of the DM is 50% at 700 nm and 95.5% at 780 nm.

The atoms decay from the high-lying odd-parity level to ground or metastable levels, the wavelength of the fluorescence is completely different from those of the pump and probe lasers. In order to reduce the background caused by the scattered light of the pump and probe beams, another cut filter is prepared. It blocks the light whose wavelength is longer than 610 nm (denoted as SWPF610).

In order to check the performance of the optical double-resonance technique, some $E_1$ transitions are measured after populating the 15650.55-cm$^{-1}$ level with the locked pump laser. The measured $E_1$ transitions with optical double-resonance technique are listed in Table 7.1.

After finishing the $E_1$ measurements with optical double-resonance technique, we start the measurement of the Stark-induced $E_1$ transitions to the levels listed in Table 7.2.
7.1. EXPERIMENTS

Figure 7.1: Overview of the optical double-resonance experimental setup. The abbreviations used are as follows. PMT represents a photomultiplier tube, MCS a multichannel scaler, FPI a Fabry-Perot interferometer, $\lambda/2$ a half-wave plate, $\lambda/4$ a quarter-wave plate, DM a dichroic mirror, PBS a polarized beam splitter, AOD an acousto-optic deflector, PC a personal computer, ADC an analog-to-digital converter and DAC a digital-to-analog converter. IP1 and IP2 denote an upstream and downstream interaction points, respectively.

Table 7.1: Measured $E_1$ transitions with ODR technique. The abbreviation Config. denotes the configuration.

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>High-lying even-parity level</th>
<th>Energy (cm$^{-1}$)</th>
<th>Config. and $J$</th>
</tr>
</thead>
<tbody>
<tr>
<td>765.6</td>
<td></td>
<td>28708.20</td>
<td>$4f^66s7s^1F_1$</td>
</tr>
<tr>
<td>746.8</td>
<td></td>
<td>29037.25</td>
<td>$4f^66s7s^9F_2$</td>
</tr>
<tr>
<td>745.2</td>
<td></td>
<td>29066.02</td>
<td>$4f^66s^2J = 2$</td>
</tr>
</tbody>
</table>

Table 7.2: Measured Stark-induced $E_1$ transitions with ODR technique. The abbreviation Config. denotes the configuration.

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>High-lying odd-parity level</th>
<th>Energy (cm$^{-1}$)</th>
<th>Config. and $J$</th>
</tr>
</thead>
<tbody>
<tr>
<td>798.7</td>
<td>$4f^55d6s^2J = 0$</td>
<td>28168.22</td>
<td></td>
</tr>
<tr>
<td>794.5</td>
<td>$4f^55d6s^2J = 1$</td>
<td>28233.08</td>
<td></td>
</tr>
<tr>
<td>765.9</td>
<td>$4f^55d6s^2J = 1$</td>
<td>28704.25</td>
<td></td>
</tr>
</tbody>
</table>
Table 7.3: Branching ratio of 28708.20-cm\(^{-1}\) level.

<table>
<thead>
<tr>
<th>Wavelength</th>
<th>28708.20 cm(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>300–400 nm</td>
<td>(\leq 0.0020)</td>
</tr>
<tr>
<td>400–500 nm</td>
<td>0.0055(15)</td>
</tr>
<tr>
<td>500–600 nm</td>
<td>0.0745(42)</td>
</tr>
<tr>
<td>600–700 nm</td>
<td>0.450(32)</td>
</tr>
<tr>
<td>700–800 nm</td>
<td>0.470(26)</td>
</tr>
<tr>
<td>(\geq 800) nm</td>
<td>(\leq 0.020)</td>
</tr>
</tbody>
</table>

7.2 Results and Analysis – \(E_1\) Transition –

7.2.1 Preliminary Checks of ODR

The fluorescence was observed for all the transitions to the levels listed in Table 7.1. Typical spectra of 765.6-nm transition are shown in Fig. 7.2. They are the spectra (a) without optical filter and (b) with the filter of SWPF610. Thus the wavelength of fluorescence was shorter than those of pump and probe beams. The spectrum when the pump laser was blocked is shown in Fig. 7.2 (c), and the peak had disappeared. We could check that the fluorescence was really owing to the optical double-resonance from these results.

We also use the peak intensity of 765.6-nm \(E_1\) transition as a reference comparing the transition intensities with that of Stark-induced \(E_1\) transitions since this \(E_1\) transition has the same lower level at 15650.55 cm\(^{-1}\) of the Stark-induced \(E_1\) transitions. Although the oscillator strength of the 765.6-nm \(E_1\) transition is unknown, this transition which is the type of \(4f^66s6p-4f^66s7s\) is useful for the comparison.

7.2.2 Estimation of Branching Ratio

Typical spectra of 765.6-nm transition with several wavelength windows are shown in Fig. 7.3. The fluorescence was observed with 400–500-nm, 500–600-nm, 600–700-nm and 700–800-nm bandpass windows. We estimate the decay branching ratio of 28708.20-cm\(^{-1}\) level for each optical filter as shown in Table 7.3. In this estimation, we take into account the transmissions of electrodes.

The decay process of the high-lying even-parity level is complicated. It seems to be dominated by two-step \(E_1\) processes via an intermediate odd-parity level. The lowest odd-parity level lies at 13796.36 cm\(^{-1}\) so that the fluorescence from the first decay \(E_1\) transition has the wavelength longer than 670 nm (as shown in Fig. 7.4 (a)). If the fluorescence with the wavelength shorter than 500 nm is emitted, the second decay \(E_1\) transition occurs from the level higher than 20000 cm\(^{-1}\) (as shown in Fig. 7.4 (b)). As the second decay processes follow the first decay processes, the whole probability of the second decay transitions becomes smaller than that of the first decay. It is consistent with the experimental results that the branching ratios of the decay with the wavelength of 600–800 nm are larger than those of 300–600 nm.

Since the 765.6-nm \(E_1\) transition is used as a reference, it is necessary to reconstruct the intensity of 765.6-nm \(E_1\) transition from the fluorescence intensity. In order to obtain the population of 28708.20-cm\(^{-1}\) level, we need to simulate the bare branching ratio of the level under several assumptions. The decay process is assumed as \(|i^+\rangle \rightarrow |n^-\rangle \rightarrow |f^+\rangle\). The initial level \(|i^+\rangle\) is the 28708.20-cm\(^{-1}\) level. The intermediate levels \(|n^-\rangle\) are the odd-parity levels
Figure 7.2: Typical spectra of 765.6-nm transition with optical double resonance; (a) without optical filter and (b) with the filter of SWPF610. (c) The pump laser was blocked.
Figure 7.3: Spectrum of 765.6-nm transition with several wavelength windows; (a) with 300–400-nm window, (b) with 400–500-nm window, (c) with 500–600-nm window, (d) with 600–700-nm window, (e) with 700–800-nm window and (f) with window longer than 800 nm. The fluorescence was observed in (b), (c), (d) and (e).
Figure 7.4: Decay processes of 28708.20-cm\(^{-1}\) level: (a) long wavelength (b) short wavelength.

whom's energy are less than 28708.20 cm\(^{-1}\). The final levels \(|f^+\rangle\) are ground and metastable levels. As the level \(|i^+\rangle\) has the total angular momentum \(J = 1\), the levels \(|n^-\rangle\) and \(|f^+\rangle\) must have \(J = 0, 1, 2\) and \(J = 0, 1, 2, 3\), respectively. 75 intermediate levels and 15 final levels satisfy such restrictions.

We regard that the detected fluorescence is not originated from the stimulated emission but from the spontaneous emission. The transition probability of spontaneous emission between upper level \(|u\rangle\) and lower level \(|l\rangle\) is given as an Einstein’s coefficient,

\[
A_{ul} = \frac{2\pi e^2}{m_e c \epsilon_0 \lambda^2} g_l f_{ul} = \frac{16\pi^3}{3\hbar \epsilon_0 \lambda^3} |\langle l|d_z|u\rangle|^2, \tag{7.1}
\]

where \(c\) speed of light in vacuum, \(\epsilon_0\) permittivity in a vacuum, \(g_{ul,l}\) the degeneracies, \(f_{ul}\) atomic oscillator strength and \(\hbar\) Planck constant. The transition probabilities are calculated for all possible transitions of \(|i^+\rangle \rightarrow |n^-\rangle\) and \(|n^-\rangle \rightarrow |f^+\rangle\). All the dipole matrix elements are assumed to be same as 0.1 \(ea_0\).}

The calculated branching ratios of the bare event for each bandpass window are listed in Table 7.4. Comparing with the experimental result listed in Table 7.3, the tendency between them is similar except for the wavelength region longer than 800 nm. However we should take into account the lifetime. The transitions in this wavelength region, in general, have long lifetime. During the interval of lifetime, atoms flow downstream. The detection efficiency at IP1 decreases as the light source leaves the center of mirror where is the most efficient for the detection. The decline of detection efficiency due to the off-center of light source is described in Appendix A. The lifetime is gives by the inverse of \(A_{ul}\). Taking into account the simulated detection efficiency and lifetime, the branching ratios are calculated as listed in Table 7.4. Here we assume the atomic-beam velocity to be 411 m/s corresponding to the temperature of \(T=1200\) K. Although, it becomes more similar to the experimental result, the discrepancy still remains.
Table 7.4: Simulated branching ratio of 28708.20-cm\(^{-1}\) level. All the E1 dipole matrix elements are assumed to be 0.1 \(e_0\).

<table>
<thead>
<tr>
<th>Wavelength</th>
<th>Bare event</th>
<th>Simulated signal</th>
</tr>
</thead>
<tbody>
<tr>
<td>300–400 nm</td>
<td>0.00098</td>
<td>0.00</td>
</tr>
<tr>
<td>400–500 nm</td>
<td>0.030</td>
<td>0.0047</td>
</tr>
<tr>
<td>500–600 nm</td>
<td>0.13</td>
<td>0.10</td>
</tr>
<tr>
<td>600–700 nm</td>
<td>0.28</td>
<td>0.36</td>
</tr>
<tr>
<td>700–800 nm</td>
<td>0.29</td>
<td>0.37</td>
</tr>
<tr>
<td>(\geq) 800 nm</td>
<td>0.27</td>
<td>0.16</td>
</tr>
</tbody>
</table>

Figure 7.5: Linearity check of E1 intensity versus the probe laser power.

7.2.3 Linearity Check of E1 Intensity

In order to use the E1 intensity as a reference for the Stark-induced E1 intensity, it is necessary to confirm that the peak intensity was not saturated owing to the probe laser power. We measured the linearity of peak intensity of 765.6-nm transition against the probe laser power as shown in Fig. 7.5. The horizontal axis is the probe laser power measured below IP1. As a result, we could check that the peak intensity was not saturated.

7.3 Results and Analysis – Stark-induced E1 Transition –

We observed the peaks of 794.5-nm transition clearly. For 798.7-nm and 765.9-nm transitions, we found that the fluorescent signals were smaller than the background in our apparatus. This means, as these transitions have the common lower level, that the E1 amplitude from the 15639.80-cm\(^{-1}\) level to 28233.08-cm\(^{-1}\) level is larger than the others, provided that the Stark mixing on the upper level is negligible.
7.3. RESULTS AND ANALYSIS – STARK-INDUCED E1 TRANSITION –

Table 7.5: Branching ratio of 28233.08-cm\(^{-1}\) level.

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Branching ratio (×10(^{-4}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>300–400</td>
<td>0.188(31)</td>
</tr>
<tr>
<td>400–500</td>
<td>–</td>
</tr>
<tr>
<td>500–600</td>
<td>0.812(70)</td>
</tr>
<tr>
<td>600–700</td>
<td>–</td>
</tr>
<tr>
<td>700–800</td>
<td>–</td>
</tr>
<tr>
<td>$\geq$ 800</td>
<td>–</td>
</tr>
</tbody>
</table>

7.3.1 Estimation of Branching Ratio

Typical spectra of 794.5-nm transition with several wavelength windows are shown in Fig. 7.6. We estimate the decay branching ratio of 28233.08-cm\(^{-1}\) level for each optical filter as shown in Table 7.5. The upper limits for the wavelengths where the fluorescence was not observed are not determined since the fluctuation of background levels were much larger than the observed signals. The fluorescence was observed with 300–400-nm and 500–600-nm bandpass windows. The tendency of decay wavelength is consistent with that in the electric discharge experiments.

7.3.2 Electric Field Dependence of Intensity

The spectra for 794.5-nm transition for various electric-field strengths are shown in Fig. 7.7. The polarization of the probe laser was set perpendicular to the direction of the electric field to observe the $\sigma$ component only. Because of the high frequency jitter of the pump laser, the population of the 15650.55-cm\(^{-1}\) level fluctuated during the measurement. Then the lineshapes of the peaks were affected by the fluctuation and slightly deviated from normal Lorentzian shape (as in the case of 17.05 kV/cm data). The data having the FWHM deviated by more than one standard deviation were rejected as shown in Fig. 7.8.

In Fig. 7.9, each peak intensity was normalized to the reference 765.6-nm transition intensity and plotted as a function of electric field strength $\mathcal{E}$. Although the data points are somewhat scattered caused by the fluctuation of the population of the 15650.55-cm\(^{-1}\) level, it is obvious that the peak intensity increases with the electric field. The intensity should be expressed by the quadratic polynomial of the electric field strength $\mathcal{E}$: The quadratic term arises from the Stark-induced E1 amplitude, whereas the linear term arises from the interference of the Stark-induced E1 amplitude with the M1 or E2 amplitude. The constant term represents the M1 or E2 transition. Since no peaks were found without electric field, the data points were fitted with the quadratic polynomial passing through the origin:

$$ r(\mathcal{E}) = r_1 \mathcal{E} + r_2 \mathcal{E}^2. $$ (7.2)

As a result, we found the coefficients of linear and quadratic terms to be

$$ r_1 = 2(4) \times 10^{-4} \text{ cm/kV}, $$ (7.3)
$$ r_2 = 8(3) \times 10^{-5} \text{ cm}^2/\text{kV}^2, $$ (7.4)

respectively.
Figure 7.6: Spectrum of 794.5-nm transition with several wavelength windows; (a) with 300–400-nm window, (b) with 400–500-nm window, (c) with 500–600-nm window, (d) with 600–700-nm window, (e) with 700–800-nm window and (f) with window longer than 800 nm. The fluorescence was observed in (a) and (c).
7.3. RESULTS AND ANALYSIS – STARK-INDUCED E1 TRANSITION –

Figure 7.7: Spectra of 794.5-nm transition for various electric-field strengths $E$. The polarization of the probe laser was set perpendicular to the electric field to observe the $\sigma$ component only.

Figure 7.8: Variation of FWHM of 794.5-nm transition peak for every data. The data having the FWHM deviated by more than one standard deviation, which indicated as a shadowed area, were rejected.
CHAPTER 7. OPTICAL DOUBLE-RESONANCE EXPERIMENTS

Figure 7.9: Electric field dependence of 794.5-nm transition peaks. Each peak intensity (794.5-nm transition) normalized to reference intensity (765.6-nm transition) as a function of $E$.

7.3.3 Determination of $\alpha_2$

In order to check the contribution of Stark mixing on the upper level of 794.5-nm transition, Stark splitting was measured. Rotating the laser polarization with the half-wave plate, the $\pi$ and $\sigma$ components were identified clearly as shown in Fig. 7.10. The tensor polarizability $\alpha_2$ has been determined to be -213.5(45) kHz/(kV/cm)$^2$ for 28233.08-cm$^{-1}$ level. Therefore it is necessary to take into account the contribution of Stark mixing on 28233.08-cm$^{-1}$ level.
7.3. RESULTS AND ANALYSIS – STARK-INDUCED E1 TRANSITION –

Figure 7.10: Stark splitting of 794.5-nm transition for different linear. Applied electric field was 17.59 kV/cm: (a) $\pi$ field, (b) Mixed field between $\pi$ and $\sigma$ fields and (c) $\sigma$ field. (d) The level scheme and corresponding transitions.
Chapter 8

Discussion

8.1 Isotope Dependence of $\alpha_2$

The level scheme around the low-lying odd-parity levels of $4f^66s6p^7G_{1-3}$ is shown in Fig. 8.1. Since the 15650.55-cm$^{-1}$ level has close-lying opposite-parity neighbor where the energy separation is only 10.75 cm$^{-1}$, $\alpha_2$ of this level is larger than those of other levels. The energy separation between the 16116.42-cm$^{-1}$ level and the closest neighbor is 161.18 cm$^{-1}$. For the 16748.30-cm$^{-1}$ level, the close-lying opposite-parity neighbor is not found. Moreover, we can clearly observe the isotope dependence of $\alpha_2$ in the 15650.55-cm$^{-1}$ level for the first time. This phenomenon is very interesting and worth discussing.

As seen in Eq. (2.38), $\alpha_2$ is proportional to the sum of squared matrix element and reciprocal of the energy separation. In case of the 15650.55-cm$^{-1}$ level, we assume that the contributions of the opposite-parity neighbors except for 15639.80-cm$^{-1}$ level are negligible. One possible reason for the isotope dependence of $\alpha_2$ is the difference of the energy separation caused by the level isotope shifts. The level isotope shifts of the 15650.55-cm$^{-1}$ and 15639.80-cm$^{-1}$ levels would yield the difference of the energy separation as shown in Fig. 8.2. Since the energy separation between the center of the fine structure levels is very small, the value of $\alpha_2$ is sensitive to the little difference of the energy separations.

However, we have no information about the isotope shift of the $M1$ transition to the 15639.80-cm$^{-1}$ level. We assume following situations. The matrix element $\langle 15650.55|d_z|15639.80 \rangle$ is equal for all isotopes. The level isotope shift of 15639.80-cm$^{-1}$ level is a little bit wider than that of 15650.55-cm$^{-1}$ level for each isotope. The level of $^{154}$Sm lies lower than that of $^{144}$Sm. Here we estimate the difference of the energy separation as

\[
\frac{d\alpha_2}{\alpha_2} = -\frac{d(\Delta E)}{\Delta E},
\]

\[
|d(\Delta E)| = \left| \frac{d\alpha_2}{\alpha_2} \frac{\Delta E}{\Delta E} \right| = \left| \frac{\alpha_2(154) - \alpha_2(144)}{\alpha_2} \Delta E \right| = \left| \frac{-546.40 - (-549.63)}{-547.81} \times 10.75 \times 30 \right| \text{GHz} 
\approx 2.0 \text{ GHz.}
\]
8.1. ISOTOPE DEPENDENCE OF $\alpha_2$

\[ 1654.60 \text{ cm}^{-1} 4f^55d6s \, D_i \]

\[ 15955.24 \text{ cm}^{-1} 4f^55d6s \, G_i \]
\[ 15914.55 \text{ cm}^{-1} 4f^56s^2 \, D_i \]
\[ 15793.68 \text{ cm}^{-1} 4f^55d6s \, F_i \]
\[ 15639.80 \text{ cm}^{-1} 4f^55d6s \, G_i \]

Even Odd

\[ 15650.55 \text{ cm}^{-1} 4f^56s6p \, G_i \]

\[ 16748.30 \text{ cm}^{-1} 4f^66s6p \, G_i \]

\[ 16166.42 \text{ cm}^{-1} 4f^66s6p \, G_i \]

\[ 14783.51 \text{ cm}^{-1} 4f^55d6s \, D_i \]
\[ 14550.50 \text{ cm}^{-1} 4f^55d6s \, P_i \]

Figure 8.1: Close-lying even-parity neighbors around low-lying levels.

Figure 8.2: difference of the energy separation caused by the level isotope shifts.
If the level isotope shift of the 15639.80-cm\(^{-1}\) is 2-GHz wider than that of the 15650.55-cm\(^{-1}\) level, the observed isotope dependence of \(\alpha_2\) will be realized.

Although the estimated isotope shift of 15639.80-cm\(^{-1}\) level is quite possible, there is another reason that difference of the matrix elements yields the isotope dependence of \(\alpha_2\). Although it should be taken into account, there is little information about the matrix element. One possible explanation is that the difference of wave function for each isotope is caused by the shape of the nucleus. The nucleus of \(^{144}\text{Sm}\) has a spherical shape while that of \(^{154}\text{Sm}\) has a deformed shape.

If the difference of the matrix element exists, the isotope dependence of \(\alpha_2\) possibly offer a very good criteria to refine the calculation of the wave function.

### 8.2 Contributor to Stark-induced \(E1\) Transition

#### 8.2.1 Large \(E1\) Amplitude

The intensity of Stark-induced \(E1\) transition is affected by two factors; the amplitude of the \(E1\) transition and the Stark mixing coefficient. We denote the 15650.55-cm\(^{-1}\), 15639.80-cm\(^{-1}\) and 28233.08-cm\(^{-1}\) levels as |1\>, |2\> and |3\>, respectively (Fig. 8.3). The level schemes around the high-lying odd-parity levels are shown in Fig. 8.4. As we know, the nearest even-parity level of the 28233.08-cm\(^{-1}\) level is at 28708.20 cm\(^{-1}\) \(^{42}\), we take the 28708.20-cm\(^{-1}\) level as |4\>.

Assuming that a level |\(i\)> with the energy \(E_i\) is mixed with an opposite-parity level |\(j\)> with \(E_j\) caused by the dominant Stark mixing under the electric field \(\mathcal{E}\), the Stark mixing coefficient \(\delta_{ij}\) is written as

\[
\delta_{ij} = \frac{(\langle i|H_{\text{Stark}}|j\rangle)}{E_i - E_j} = \frac{(\langle i|d_z|j\rangle)}{E_i - E_j}\mathcal{E}.
\]

(8.1)

Under the same assumption, the dipole matrix element in Eq. (8.1) is expressed with \(\alpha_2\) as shown in Eq. (2.40). And then, we have

\[
|\delta_{ij}| = \begin{cases} 
\frac{3}{2} \frac{(J_i^2 - m_i^2)}{J_i(2J_i - 1)} \frac{\alpha_2}{(E_i - E_j)} \mathcal{E} & \text{for } J_j = J_i - 1 \\
\frac{3}{2} \frac{m_i^2}{J_i(2J_i - 1)} \frac{\alpha_2}{(E_i - E_j)} \mathcal{E} & \text{for } J_j = J_i \\
\frac{3}{2} \frac{(J_j + 1)^2 - m_j^2}{J_j(2J_j - 1)} \frac{\alpha_2}{(E_i - E_j)} \mathcal{E} & \text{for } J_j = J_i + 1,
\end{cases}
\]

(8.2)

where \(J_i\) and \(J_j\) are the electronic angular momenta of levels |\(i\)> and |\(j\)>, respectively, and \(m_i\) is the projection of \(J_i\) along quantization axis.

The intensity ratio of Stark-induced \(E1\) transition to the reference transition, \(r_2\), is expressed as

\[
r_2 = \frac{|\delta_{12}A_{23} + \delta_{34}A_{14}|^2}{|A_{14}|^2} = \delta_{12}^2 \left| \frac{A_{23}}{A_{14}} \right|^2 + \delta_{34}^2 + 2\text{Re} \left( \delta_{12}^* \delta_{34} \frac{A_{23}^2}{A_{14}^2} \right),
\]

(8.3)

where \(A_{14}\) and \(A_{23}\) are the \(E1\) amplitude from the level |1\> to level |4\> and that from the level |2\> to level |3\>, respectively.
8.2. CONTRIBUTOR TO STARK-INDUCED E1 TRANSITION

Figure 8.3: Contributors to the 794.5-nm Stark-induced E1 transition. $\delta_{12}$ and $\delta_{34}$ represent the Stark mixing coefficients on the level $|1\rangle$ and $|3\rangle$, respectively. $A_{14}$ and $A_{23}$ are the $E1$ amplitude from the level $|1\rangle$ to level $|4\rangle$ and that from the level $|2\rangle$ to level $|3\rangle$, respectively.

Figure 8.4: Close-lying even-parity neighbors around high-lying levels.
The value of $\delta_{12}$ is derived by substituting Eq. (2.41) to Eq. (8.1) with $\alpha_2 = -561.7(11)$ kHz/(kV/cm)$^2$ [34] and $\Delta E=10.75$ cm$^{-1}$;

$$\delta_{12} = 1.617(25) \times 10^{-3} \text{ cm/kV } \mathcal{E}. \quad (8.4)$$

The value of $\delta_{34}$ is similarly derived with $\alpha_2 = -213.5(45)$ kHz/(kV/cm)$^2$ and $\Delta E=475.12$ cm$^{-1}$;

$$\delta_{34} = 1.499(16) \times 10^{-4} \text{ cm/kV } \mathcal{E}. \quad (8.5)$$

The errors of $\delta_{12}$ and $\delta_{34}$ come from the uncertainties of $\alpha_2$ and the energy separation. We assume the uncertainty of the energy separation to be 10 GHz by considering the isotope shift of Sm. As the value of $r_2$ is obtained in Eq. (7.4) to be $8(3) \times 10^{-5}$ cm$^2$/kV$^2$ $\mathcal{E}^2$.

Substituting $\delta_{12}$, $\delta_{34}$ and $r_2$ to Eq. (8.3), we can deduce

$$\left| \frac{A_{23}}{A_{14}} \right| = 5(1). \quad (8.6)$$

Even if the level $|3\rangle$ were mixed with the second or third nearest even-parity levels, $|A_{23}/A_{14}|$ would be larger than the above value. This does not change the fact that $A_{23}$ is estimated to be anomalously large. This fact suggests that the transition from level $|1\rangle$ to level $|3\rangle$ provides a prospective means for a PNC measurement.

### 8.2.2 Unknown Close-lying Even-parity Level

Since we do not have enough knowledge at present for even-parity levels in the region of 20000–30000 cm$^{-1}$, a possibility that unknown close-lying even-parity level induces a large Stark mixing to the level $|3\rangle$ remains. The result obtained in the electric discharge experiment suggests a possibility that the level $|3\rangle$ includes the configurations of $4f^65d6p$ or $4f^66s6p$ in addition to the leading configuration of $4f^55d6s^2$. If this is the case, the unknown even-parity level $|4\rangle$ having the configurations of $4f^65d6s$, $4f^66s^2$ or $4f^66s7s$ could admix with the level $|3\rangle$, and the $E1$ amplitude between the levels $|1\rangle$ and $|4\rangle$, $A_{14\prime}$, could be allowed. In this case, Eq. (8.3) can not be solved uniquely: For instance, $\delta_{34\prime} \simeq 7.3 \times 10^{-3}$ is deduced by assuming that $A_{14} \simeq A_{23} \simeq A_{14\prime}$. Substituting $\alpha_2$ and $\delta_{34\prime}$ to Eq. (8.2), we find that the level $|4\rangle$ with $J$ must lie at 28233.08 cm$^{-1}$ +6 MHz ($J=0$), −6 MHz ($J=1$) or +24 MHz ($J=2$). If such extremely close-lying level existed, the peaks would have been observed in the measurements of transition to the level $|3\rangle$.

### 8.3 Feasibility for Measuring the PNC Effects with ODR

#### 8.3.1 Improvement of Signal-to-noise Ratio

We discuss here the signal-to-noise ratio, $S/N$, for the Stark-induced $E1$ transition. The signal $S$ is a height of Stark-induced $E1$ transition peak. The noise $N$ is a DC offset under the peak. In the $E1$ measurement for the transition from the 0-cm$^{-1}$ to 15650.55-cm$^{-1}$ levels, the observed $S$ were 50000 counts/20ms while the $N$ were 500 counts/20ms. The $S/N$ of Stark-induced $E1$ transition in the one-step excitation was estimated from the $S/N$ of measured $E1$ transition. On the other hand, we have directly observed the Stark-induced $E1$ transition with optical-double resonance technique and can extract the measured $S/N$. The $S/N$’s of Stark-induced $E1$ transition peak with each filter windows are listed in Table 8.1.
8.3. FEASIBILITY FOR MEASURING THE PNC EFFECTS WITH ODR

Table 8.1: Signal-to-noise ratio of Stark-induced $E_1$ transition peak. $S$ and $N$ are given in unit of counts/20 ms. The sources of $N$ are also listed. SL indicates the scattered light originated from incident laser. TR indicates the thermal radiation from atomic-beam oven. EN indicates the electric noise.

<table>
<thead>
<tr>
<th>Filter</th>
<th>$S$</th>
<th>$N$</th>
<th>$S/N$</th>
<th>Source of $N$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1-step excitation</td>
<td>0.13 $\varepsilon^2$</td>
<td>500</td>
<td>$2.6 \times 10^{-4} \varepsilon^2$</td>
<td>SL, TR, EN</td>
</tr>
<tr>
<td>Optical double resonance</td>
<td>0.16 $\varepsilon^2$</td>
<td>5</td>
<td>$3.2 \times 10^{-2} \varepsilon^2$</td>
<td>EN</td>
</tr>
<tr>
<td>300–400 nm</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>500–600 nm</td>
<td>0.68 $\varepsilon^2$</td>
<td>20</td>
<td>$3.4 \times 10^{-2} \varepsilon^2$</td>
<td>TR, EN</td>
</tr>
<tr>
<td>$\leq$ 610 nm</td>
<td>0.89 $\varepsilon^2$</td>
<td>30</td>
<td>$3.0 \times 10^{-2} \varepsilon^2$</td>
<td>TR, EN</td>
</tr>
</tbody>
</table>

The obtained $S/N$ is 100 times larger that estimated in the one-step $E_1$ experiments. This results obviously shows the advantage of optical-double resonance measurement against one-step excitation measurement. The sources of $N$ are also listed in Table 8.1. In the one-step $E_1$ experiments, the most part of $N$ originates from the scattered light of incident laser (SL). Other sources of $N$ are the thermal radiation from the atomic-beam oven (TR) and the electric noise (EN). Contrarily, the sources of $N$ in the optical double-resonance experiments are TR an EN. The reduction of $N$ is due to the blocking of SL with optical filter, which we have just expected during the planning.

Moreover, the $S$ is larger than the one expected in the one-step $E_1$ experiments. As mentioned in previous section, it is possibly caused by the admixture of large $E_1$ amplitude or the contribution of high-lying level to the Stark mixing. There is a possibility that we can find more suitable level providing larger $S$ by investigating other high-lying levels.

8.3.2 Required Improvement in ODR Experiments

In this study, we have performed the observation of Stark-induced $E_1$ transition with optical-double resonance technique for the first time. Several problems have been clarified in the experiment.

Large fluctuation of peak intensity may make off the asymmetry of transition intensity: The fluctuation was as large as the 30% of peak intensity in the worst case. This was due to the high frequency jitter of the pump laser which could not be stabilized in the software locking system. The emitting rate of the fluorescence of Sm is so slow ($\sim 10 \mu s$) that the integration of fluorescent signals is necessary to generate the error signal to be used in the feedback system. Using faster frequency reference such as FM-sideband method with high-stabilized FPI, the high frequency jitter will be reduced.

In Ref. [54], we had discussed the required precision for measuring the asymmetry in order to obtain the desired precision of $Q_W$. In spite of the enlargement of $S/N$ from previous study, the statistics still demands extremely large accumulation of data in order to obtain $Q_W$ with 0.1% precision. Thus we have to improve our system more to obtain large $S$. Possible improvements obtaining large $S$ were discussed in our previous studies [48,54].

The most drastic improvement is to introduce a power-buildup cavity (PBC) whose effectiveness was proved in the successful Cs experiment [5]. High-finess cavity makes it possible to amplify the laser intensity at interaction point with atomic beam. We can expect the multiplication of $S$ as much as the finess of PBC, realistically about $10^4$. However, in the optical double-resonance experiments, we have to introduce the pump laser away from PBC by tilting
its incident angle slightly. Moreover, the laser frequency stabilization is required for the probe laser in order to tune its frequency to resonance on PBC.

With the buildup of probe laser, it is necessary to populate the 15650.55-cm$^{-1}$ level more. Since the pump laser power was considerably lower than that of probe laser in this study, more powerful one is required. Stabilizing the pump laser leads to high population of 15650.55-cm$^{-1}$ level. This also gives the reduction of high frequency jitter, which is already described.

A powerful but rather simple improvement is to increase the atomic-beam intensity. Reducing the distance $d$ between the orifice of crucible and IP1 is effective as the atomic-beam intensity is proportional to $1/d^2$. In addition, we can devise a more directional and high intensity beam with the help of the capillary array at the exit [5,67].
Chapter 9

Conclusions

Spectroscopic study of Sm has been performed as a preliminary investigation for the PNC measurements. The systematic study of $4f^6s^6p^7G$ term in Sm was performed by atomic-beam laser spectroscopy. Hyperfine splittings and isotope shifts were measured for five $E1$ transitions from $4f^6s^2F_{2-6}$ to $4f^6s^6p^7G_{2-6}$ levels without external field. Stark spectroscopy was performed for seven $E1$ transitions from $4f^6s^2F_{0-6}$ to $4f^6s^6p^7G_{1-6}$ levels under external electric field. Zeeman spectroscopy was performed for five $E1$ transitions from $4f^6s^2F_{2-6}$ to $4f^6s^6p^7G_{2-6}$ under external magnetic field.

We have newly determined the hyperfine coupling constants $A$ and $B$ of $4f^6s^6p^7G_{2-6}$ levels for $^{149}$Sm and $^{147}$Sm. King plot analysis have been applied to the obtained isotope shifts. The specific mass shift and field shift have been separated from the observed isotope shifts. The electron density difference at the nucleus $\Delta|\psi(0)|^2_1$ and nuclear parameter $\lambda$ have been derived for five transitions.

From the Stark splittings, the tensor polarizabilities $\alpha_2$ of $15650.55$-cm$^{-1}$ ($4f^6s^6p^7G_1$), $16116.42$-cm$^{-1}$ ($4f^6s^6p^7G_2$) and $16748.30$-cm$^{-1}$ ($4f^6s^6p^7G_3$) levels have been determined to be $-548(12)$, $-112.5(24)$ and $119.8(26)$ kHz/(kV/cm)$^2$, respectively. For the other levels of $4f^6s^6p^7G_{4-6}$, Stark splittings were not resolved in our experimental resolution. We have found that the $\alpha_2$ of $15650.55$-cm$^{-1}$ level, which has close-lying opposite-parity neighbor, is considerably larger than those of others. Moreover, the isotope dependence of $\alpha_2$ has been clearly observed for the $15650.55$-cm$^{-1}$ level for the first time. We have two possible explanations for this phenomenon. One is that the $\alpha_2$ is affected by the difference of the energy separation caused by the isotope shift of relevant levels. Another is that the $\alpha_2$ reflects the difference of wave function caused by the shape of the nucleus for each isotope. If the latter is the case, the isotope dependence of $\alpha_2$ offers a good criteria to refine the calculation of the wave function.

The gyromagnetic ratios $g$-values have been determined for five levels of $4f^6s^6p^7G_{2-6}$ from the Zeeman splittings. The precisions of these values have been improved by several orders of magnitude compared with the reference values. These results would contribute to the improvement in the accuracy of the atomic wave function.

In the next stage of the experiments, we have observed the Stark-induced $E1$ transitions with optical double-resonance technique. We have performed electric discharge experiments in order to search suitable high-lying odd-parity levels. Three high-lying levels at $28168.22$ cm$^{-1}$, $28233.08$ cm$^{-1}$ and $28704.25$ cm$^{-1}$ have been chosen. While the pump laser was locked to the $E1$ transition from $0$-cm$^{-1}$ to $15650.55$-cm$^{-1}$ levels, the probe laser was tuned to the parity
forbidden transitions under electric field. In the probe transition from the 15650.55-cm$^{-1}$ to 28233.08-cm$^{-1}$ levels ($4f^55d6s^2 J = 1$), we have observed the Stark-induced $E_1$ transition. The observed peak was anomalously large compared with the $E_1$ peak measured as a reference. Our experimental results are explained by either (1) anomalously large $E_1$ amplitude from the 15639.80-cm$^{-1}$ level to the 28233.08-cm$^{-1}$ level or (2) existence of unknown close-lying level mixed with the 28233.08-cm$^{-1}$ level. The letter suggests that the transition from the 15650.55-cm$^{-1}$ to 28233.08-cm$^{-1}$ levels provides a prospective means for a PNC measurement.

The signal-noise-ratio of the observed Stark-induced $E_1$ transition has been improved by about 100 times from that estimated in the one-step $E_1$ experiments. This improvement is due to the background reduction in the optical double-resonance experiments. However, we found that further improvements were necessary to obtain precise $Q_W$.

In summary, we have studied the 15650.55-cm$^{-1}$ level as a candidate of PNC measurements. The $E_1$ and Stark-induced $E_1$ transitions connected with the 15650.55-cm$^{-1}$ level have been measured by atomic-beam laser spectroscopy. We have obtained several spectroscopic properties, such as hyperfine structures, isotope shift, tensor polarizability and $g$-value. The analysis of these values led to the conclusion that the 15650.55-cm$^{-1}$ level of Sm is possibly useful for the PNC measurements with optical double-resonance technique and is worth striving for increasing the spectroscopic signal intensity.
I am deeply indebted to Professor Ichita Endo for his instruction and guidance. I would like to thank Professor Takayoshi Horiguchi for his instructions on the experimental techniques. I am grateful to Professor Tohru Takahashi for his guidance and suggestions. I would like to express my gratitude to Dr. Masataka Iinuma for his advice and critical reading of this thesis.

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Appendix A

Detection Efficiency

We have simulated the detection efficiency at IP1 considering a spacial distribution and an offset of light source along the atomic beam line as shown in Fig. A.1. We have assumed a sectioned atomic beam as a light source. It is parallel to $y$-$z$ plane and has a Gaussian distribution. The $\sigma_y$ and $\sigma_z$ were assumed to be 2 mm and 0.5 mm, respectively. The light left the center of the mirror along $x$ axis and emitted the fluorescence in random direction. We judged if the fluorescence hit the detection area or not. Two types of detection were taken into account; the detection of direct light and that reflected by the mirror. The result of the simulation is shown in Fig. A.2. The velocity of atomic beam was assumed to be 411 m/s corresponding to the temperature $T=1200$ K.
Figure A.1: Simulation of detection efficiency. The light source traveled along $x$-axis.

Figure A.2: Simulated detection efficiency. The velocity of atomic beam was assumed to be 411 m/s corresponding to the temperature $T=1200$ K.
Bibliography


