Proceedings of the Korean Nuclear Society Autumn Meeting
Seoul, Korea, October 2001

Effect of Magnetic Field on Selectivity of Three-Step Photoionization

Changhwan Lim, Sipyo Rho, Kwanghoon Ko and Chuljoong Kim

Korea Atomic Energy Research Institute
150 Duckjin-dong, Yusong-ku
Taejon, Korea 305-353

Yasukazu Izawa
Institute of Laser Engineering, Osaka University
2-6 Yamada-oka, Suita
Osaka, Japan 565-0871

Abstract

Effect of magnetic field on selectivity by linearly polarized lasers was analyzed by formulating the density matrix equations. To investigate the effect of magnetic field on the selectivity of AVLIS, we proposed a general Hamiltonian for multilevel atomic system in magnetic field. The population dynamics of magnetic sublevels have been observed by solving the Liouville equation. Mixing between magnetic sublevels was observed in each state during the laser excitations when the magnetic field perpendicular to the quantization axis was applied to the atomic system. The magnetic field dependence on ionization rate of even isotopes was also discussed. In the magnetic field dependence, two ionization peaks were appeared because of the interference between Rabi and Larmor frequency during the ionization process.

The permissible intensities of magnetic field were predicted to obtain enough selectivity for the target isotopes of zirconium and gadolinium in the AVLIS process based on the polarization selection rule.

1. Introduction

In AVLIS, a target isotope is usually excited selectively by precisely tuned narrow band lasers using isotope shifts and subsequently ionized by another laser, while non-target isotopes remain in the ground state.[1,2] The isotope separation method using the isotope shifts has been successfully demonstrated in uranium enrichment. However, when the isotope shifts is small, selectivity is severely degraded. If the selectivity is only based on the isotope shifts, the feasibility of the atomic vapor isotope separation (AVLIS) process to those elements, such as zirconium or gadolinium, cannot be easily demonstrated.

As shown in the references, zirconium has small isotope shifts in comparison with that of uranium and moreover the hyperfine structure of the target odd isotopes overlaps with the transition lines of non-target even isotopes.[3,4] In order to enrich or deplete the target odd isotopes of these elements, isotope separation by the polarization selection rules have been proposed.[5,6] The polarization method, based on the intermediate state alignment, requires to choose an excitation scheme with proper quantum value of each level and combination of laser polarization.

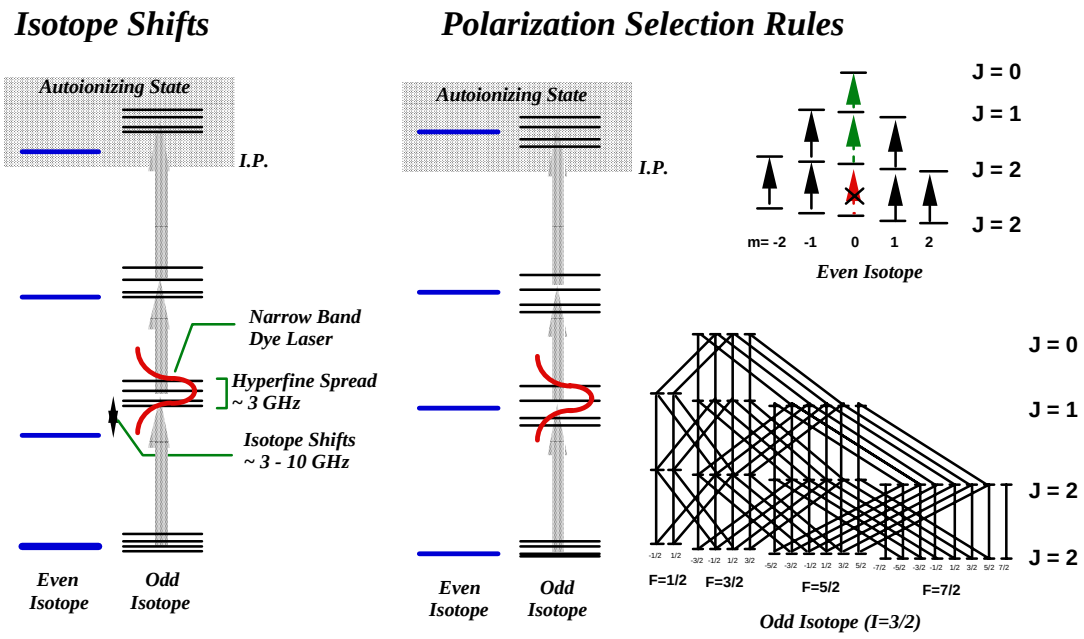

Fig.1 Three-step photo-ionization scheme based on (a) isotope shifts and (b) polarization selection rule.

When one choose an excitation scheme with proper quantum number *J* and combination of laser polarization, for example *J*=2-2-1-0 with 3 linearly polarized lasers, even isotope cannot be ionized due to the polarization selection rule, while odd isotope can be ionized through the transition between hyperfine structures. This is a generalization of the method firstly proposed by Balling and Wright for atoms with *J*=0 ground state angular momentum.[7] Figure 4.1 illustrate the two isotope separation methods based on the isotope shifts and the polarization selection rule. Since the transition *m*=0-0 between the ground and the first excited state is forbidden, the even isotopes in *m*=0 level of the ground states cannot be ionized through the second, third and forth excited state, subsequently. Bekov *et al* have applied this method to the selective photoionization of ytterbium isotope with broadband linearly polarized lasers.[8] Haynam *et al* and other research groups extended this method to gadolinium isotope separation.[5,9]

However, there are many stray magnetic field sources in a usual experimental condition; atomizer, electric power line, ion collector, or measuring equipment. Especially, in AVLIS process, atoms are vaporized by electron beam gun, where strong magnetic field is used to bend the path of electron beam.[10] The presence of a stray magnetic field, even at the low level of terrestrial magnetism, destroys the atomic state alignment prepared by the polarized lasers. As known in Hanle effect,[11,12] the magnetic field induces the precession motion of angular momentum and ionization of the even isotopes. So, selectivity is expected to be degraded by the presence of magnetic field. However, only a few study on the effect of magnetic field on laser isotope separation have been reported.

In the this paper, effect of magnetic field on AVLIS based on the polarization selection rule will be studied theoretically by solving the density matrix equations. In Sec. 2, the Hamiltonian of multilevel atomic system under the magnetic field and Liouville equation are introduced. In Sec. 3, several calculated examples of the population of motion of multilevel atomic system will be presented. And the dependence of external parameters such as ionization rate, intensity of applied magnetic field, and life time of states, on the excitation and photo-ionization will be discussed. In Sec. 4, selectivity of the three-step photo-ionization based on the polarization selection rules for zirconium and gadolinium will be discussed.

2. Hamiltonian and Liouville Equation

Figure1 illustrates a *N*-level atom with energies ω_n, interacting with *N*-1 laser fields of arbitrary strength and of frequency $\omega_{n'}$ in the arbitrary magnetic field. Each level assumed to have its own *J* value with *2J*+1 degeneracy. This degeneracy will be split into the magnetic sublevel *m* under the influence of magnetic fields. The atomic transitions are assumed to be made only through single photon processes with appropriate photon energies.

The Hamiltonian of this system can be decomposed into the sum of a atomic Hamiltonian H_a, the interaction Hamiltonian with radiation fields H_I, and the Zeeman Hamiltonian H_B which describes the interaction with the static external magnetic field, respectively,

$$H = H_a + H_I + H_B, \tag{1}$$

where

$$H_a = \sum_{n=1}^{N} \varepsilon_n \tilde{N}, \tag{2}$$

$$H_I = -\frac{1}{2}\sum_{n=1}^{N-1} |d_k| \left[\tilde{N}_k^+ E_k(t) + \tilde{N}_k^- E_k^*(t)\right],$$

$$H_B = \mu_B g_J \hat{B} \bullet \hat{J} = \beta_J \left[B_z J_z + B_+ J_+ + B_- J_-\right].$$

The number operators in H_a and H_I are defined as

$$\begin{aligned} &\tilde{N} = |n\rangle\langle n|, \\ &\tilde{N}^+ = |n\rangle\langle n+1|, \tilde{N}^- = |n+1\rangle\langle n|, \end{aligned} \tag{3}$$

and the angular momentum operator J_z, J_+, J_- in H_B is defined as

$$\begin{aligned} &J_z|nmJ\rangle = \hbar m|nmJ\rangle, \\ &J_\pm|nmJ\rangle = \hbar\sqrt{(J \mp 1)(J \pm m + 1)}|nmJ\rangle. \end{aligned} \tag{4}$$

ε_n is energy of *n*th level and d_k is dipole moment between *k*th and (*k*+1)th level. The rotating wave or resonance approximation is used in the interaction Hamiltonian H_I. The quantization axis of atomic system is assumed to be z. The radiation field $E_k(t)$ between *k*th and (*k*+1)th level can be written

$$E_k(t) = \hat{e}_z E_k^0 e^{i\omega_k t}. \tag{5}$$

It is assumed that the radiation fields is linearly polarized along the direction of z-axis. The Liouville equation for the density matrix ρ describing the system is

$$i\hbar \frac{d}{dt}\rho(t) = [H, \rho(t)]. \tag{6}$$

Time evolution of the diagonal elements of ρ obeys the equation

$$\begin{aligned} i\hbar \frac{d}{dt}\rho^{nmJ}_{n'm'J'}(t) &= (\omega_n - \omega_{n'})\rho^{nmJ}_{n'm'J'} + \hbar B_z (m-m')\rho^{nmJ}_{n'm'J'} \\ &+ \hbar B_+ \left[\sqrt{(J+1)(J-m+1)}\rho^{nm-1J}_{n'm'J'} - \sqrt{(J'-1)(J'+m'+1)}\rho^{nmJ}_{n'm'+1J'}\right] \\ &+ \hbar B_- \left[\sqrt{(J-1)(J+m+1)}\rho^{nm+1J}_{n'm'J'} - \sqrt{(J'+1)(J'-m'+1)}\rho^{nmJ}_{nm'-1J'}\right] \\ &+ \frac{1}{2} d^m_{nn+1} E_{nn+1}(t)\rho^{nmJ}_{n'+1mJ'} + \frac{1}{2} d^m_{nn-1} E_{n-1n}(t)\rho^{n-1mJ}_{n'mJ'} \\ &- \frac{1}{2} d^{m'}_{n'+1n'} E_{n'n'-1}(t)\rho^{n+1mJ}_{n'm'J'} - \frac{1}{2} d^{m'}_{n'-1n'} E_{n'-1n'}(t)\rho^{nmJ}_{n'-1m'J'} \\ &- \frac{1}{2}\left[\sum_{k,m}(R^m_{nk} + R^m_{kn'}) + 2\Gamma^m_N + \gamma_{nn'}\delta_{nn'\pm 1}\right]\rho^{nmJ}_{n'm'J'}, \end{aligned} \tag{7}$$

where n, m and J present the energy level, magnetic quantum number and the angular momentum of nth energy level, respectively, and d^m_{nn+1} can be expressed with Wigner 3j symbol and dipole moment d

$$d^m_{nn+1} = (-1)^{J_1 - m_1}\begin{pmatrix} J_1 & 1 & J_2 \\ m_1 & q & m_2 \end{pmatrix}\langle n_1 m_1 J_1 | d | n_2 m_2 J_2 \rangle. \tag{8}$$

The first term of Eq.(7) expresses the detuning and the next term expresses Zeeman splitting by magnetic field B_z. The second and third lines of Eq.(7) express coherent mixing of magnetic sublevels by the magnetic field. The fourth and fifth lines describe the coherent excitation and deexcitation from each state that is linked to state m by laser fields. R^m_{nk} express the rate per atom at which state m gains or losses population from state k by incoherent processes (i.e. spontaneous emission, radiative or collisional processes) and Γ^m_N expresses ionization rate from the highest level. At the end of Eq. (7), the laser bandwidth $1/2\,\gamma_{nn'}$ was added.

3. Population Dynamics of Multilevel Atom

At first, the population transfer between the magnetic sublevels due to the precession motion under the existence of magnetic field is considered. Here, the initial population of each magnetic sublevel at t=0 is assumed to be

$$\rho^{2-22}_{2-22}(0) = 0.2,\ \rho^{2-12}_{2-12}(0) = 0.2,\ \rho^{202}_{202}(0) = 0.0,\ \rho^{212}_{212}(0) = 0.2,\ \rho^{222}_{222}(0) = 0.2 \qquad (9)$$

The above initial population distribution can be prepared experimentally by the excitation of linearly polarized broadband short pulse laser in J=2-2 transition because the transition m=0-0 is forbidden by selection rule.

Figure 2 is the population dynamics of magnetic sublevels with a magnetic field that is perpendicular to the quantization axis z. Since there are the number of coherent couplings by lasers and magnetic field, population dynamics with a magnetic field show much complicated features than the population dynamics without magnetic fields. However, one can observe two oscillation motions in Fig. 2; the fast one is the Rabi oscillation by lasers as observed in Fig. 4.4, and the slow one is the Larmor precession by magnetic field. Even though the ionization scheme is chosen to be J=2-2-1-0 and polarization of lasers linear ones, the number of atoms are ionized through the m=0 level of the 4th state due to the population transfer by magnetic field.

Figure 3 shows the magnetic field dependence of ionization. In low magnetic field region, ionization increases as magnetic field increase because population transfer from non-zero magnetic sublevel to m=0 level by precession motion is proportional to the intensity of magnetism. On the other hand, in high magnetic field region, ionization decreases as magnetic field increases. When atoms are in the strong magnetic field, population transfers by the magnetic field between magnetic sublevels become faster than those by lasers between states. So, populations are trapped by magnetic field between magnetic sublevels and ionization is suppressed. When the Larmor frequency is comparable to the Rabi frequency, a dip appears as can be seen in Fig. 3. Two coherent motions, Rabi and Larmor frequency, can have different phase depending on the strength of magnetic field and transition probability. The phase difference of two coherent motions can induce the constructive or destructive interference, during the ionization process. The dip in Fig 3 is considered as a destructive interference between two coherent motions.

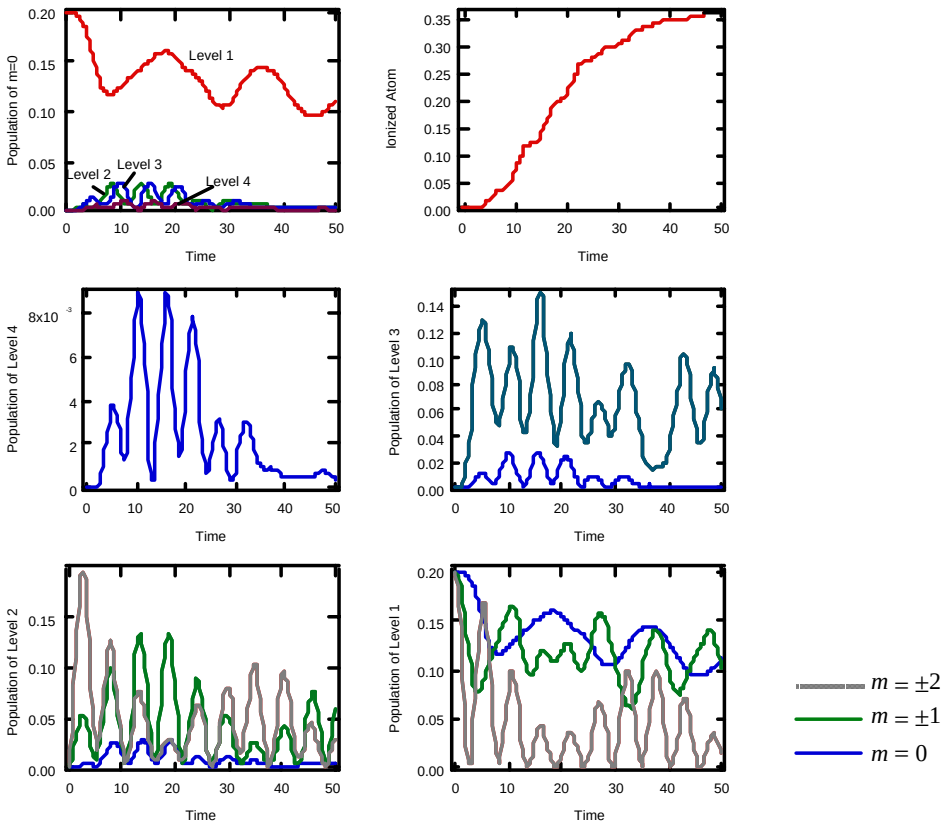

Fig. 2 Ionization as a function of delay between lasers. The sech lasers are applied to the atomic system under the static magnetic field (0.1 GHz). The pulse widths of lasers are 2π nsec and peak Rabis are 50 GHz. The spontaneous emission rate of each excited state is 10 MHz.

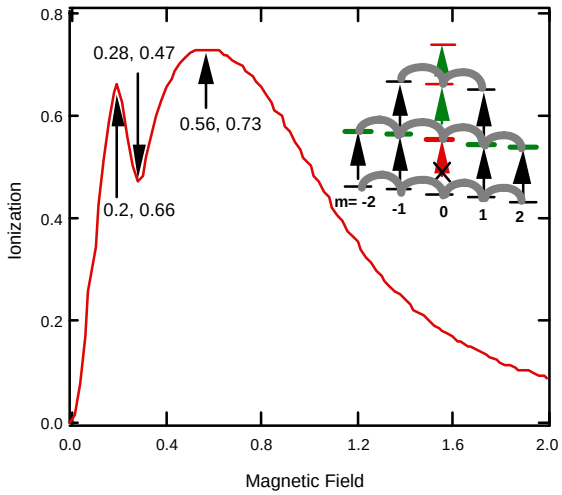

Fig. 3 Magnetic field dependence of ionization. Because of the interference between Rabi and Larmor frequency, a dip appears in this graph. Pair of Arabic numerals in graph present the magnetic field and ionization, respectively.

4. Isotope Selectivity of Multilevel Atom Based on the Polarization Selection Rule

In this section, effects of magnetic field on the isotopic selectivity for zirconium and gadolinium in AVLIS process are discussed. In the AVLIS of zirconium, ^{91}Zr depleted isotope samples are required as a products. Therefore if the process is based on the polarization selection rule, depleted zirconium products will be collected in tail as shown in Fig. 4.9. On the other hand, in case of AVLIS of gadolinium, odd gadolinium isotope ^{157}Gd is enriched in head.

Effect of magnetic field on ionization of ^{91}Zr is assumed to be quite small because the ionization pathway of odd isotope is already allowed by complicated hyperfine transition lines. It was estimated that ionization rate of ^{91}Zr was obtained to be 80% under the conditions in the previous section. This value is comparable to the theoretical maximum ionization rate of ^{91}Zr (85%). Remaining ^{91}Zr (~20%) would be stuck in tail. The ^{91}Zr abundance in tail is given by the following;

$$^{91}Zr\ abundance = \frac{(^{91}Zr, 11.2\%) \times 0.2}{(Even\ isotope, 88.8\%) \times Ionization + (^{91}Zr, 11.2\%) \times 0.2}. \quad (12)$$

On the other hand, in case of AVLIS of gadolinium, target isotope is ^{157}Gd. From the absorption spectra of gadolinium, it can be assumed that only ^{156}Gd is a non-target isotope in calculating the selectivity of gadolinium, because the absorption lines of other isotopes are far away from the hyperfine structure of ^{157}Gd.

The ^{157}Gd abundance in head can be obtained in the similar manner;

$$^{157}Gd\ abundance = \frac{(^{157}Gd, 15.68\%) \times 0.7}{(^{156}Gd, 20.47\%) \times Ionization + (^{157}Gd, 15.68\%) \times 0.7}. \quad (13)$$

The intensities of permissible magnetic field for the products are summarized in Table 1 and Table 2. External parameters for the calculation of the ionization rate of even isotopes were same to the conditions used in the calculation of magnetic field dependence and the ionization rates of odd isotope for Table 1 and 2 were calculated by CEALIS with the same conditions. The Lande *g* factor of the ground and first excited states are summarized in the parenthesis of the first column in Table 1 and 2. Lande g factor of the second and third excited states are

assumed to be 1 for the simplicity. Table 1 shows that magnetic field must be suppressed less than 5.0×10^{-5} T to obtain the ^{91}Zr abundance less than 2%. In the AVLIS of gadolinium, one can see that magnetic field must be less than 3.0×10^{-5} T to obtain the ^{157}Gd abundance more than 90% when the first excited state $^3F_1$ is used. These values are comparable to or less than the magnitude of terrestrial magnetism (0.3×10^{-5} T).

Table 1. Permissible intensity of magnetic field for ^{91}Zr depletion.

Transition Line for the 1st excitation step	Magnetic field (Gauss) 2.5% ^{91}Zr	3.0% ^{91}Zr
a^3F_2 (0.66)-z^5F_1 (0.3)	1.80	5.76
a^3F_2 (0.66)-z^5F_2 (0.67)	0.57	1.82
a^3F_2 (0.66)-z^3D_1 (0.42)	1.29	4.11
a^3F_2 (0.66)-z^3D_2 (0.96)	0.50	1.58

Table 2. Permissible intensity of magnetic field for ^{157}Gd enrichment.

Transition Line for the 1st excitation step	Magnetic field (Gauss) 90% ^{157}Gd	80% ^{157}Gd
$^9D^o_2$ (2.65)-$^7P_2$ (2.35)	0.41	0.61
$^9D^o_2$ (2.65)-$^3F_1$ (3.33)	0.29	0.43
$^9D^o_2$ (2.65)-$^3F_2$ (2.11)	0.46	0.68
$^9D^o_2$ (2.65)-$^9D_2$ (2,61)	0.37	0.55

5 Summary

In this paper, the effect of magnetic field on isotopic selectivity in AVLIS by linearly polarized lasers was analyzed by formulating the density matrix equations. To investigate the effect of magnetic field on the selectivity of AVLIS, we proposed a general Hamiltonian for multilevel atomic system in magnetic field. The population dynamics of magnetic sublevels have been observed by solving the Liouville equation. Mixing between magnetic sublevels

was observed in each state during the laser excitations when the magnetic field perpendicular to the quantization axis was applied to the atomic system. The loss rate and magnetic field dependence on ionization rate of even isotopes were also discussed. The maximum ionization rate was obtained when the ionization rate is comparable to the generalized Rabi frequency. In the magnetic field dependence, two ionization peaks were appeared because of the interference between Rabi and Larmor frequency during the ionization process.

The permissible intensities of magnetic field were predicted to obtain enough selectivity for the target isotopes of zirconium and gadolinium in the AVLIS process based on the polarization selection rule. Magnetic field should be less than 3.0×10^{-5} T to obtain the ^{157}Gd abundance more than 90% and 5.0×10^{-5} T to obtain the ^{91}Zr abundance less than 2%. From the above results, one can see that the magnetic field in AVLIS of zirconium and AVLIS of gadolinium based on the polarization selection rule must be less than the terrestrial magnetism to obtain the commercial products.

References

1. V. S. Letokhov, "Laser Isotope Separation", Nature 277 (1979), 22
2. R .C. Stern and J. A. Paisner, "Atomic Vapor Laser Isotope Separation" Lawrence Livermore National Laboratory, UCRL-93584 (1985).
3. C. Lim, K. Nomaru, and Y. Izawa, **"**Hyperfine structure constants and isotope shifts determination of ZrI by laser induced fluorescence spectroscopy ", Jpn. J. Appl. Phys., Vol. 37, No.9 pp.5049-5202 (1998); C. Lim, S. Morikawa, K. Nomaru, and Y. Izawa, "Isotope Shifts and Hyperfine Structure Constants in ZrI Measured by Laser Induced Fluorescence Spectroscopy", Technology Reports of Osaka University, Vol. 47, No. 2275, pp.71-77 (1997).
4. C. Lim and Y. Izawa, **"**Hyper fine structure and isotope shifts of high-lying levels of ZrI by laser induced fluorescence spectroscopy ", J. Opt. Soc. Am. B, Vol.10, No. 1, pp.2607-2713 (1998).
5. C. Haynam, B. Comaskey, J. Conway, J. Eggert, J. Glaser, E. Ng, J. Paisner, R. Solarz, and E. Worden, " Status of gadolinium enrichment technology at LLNL" SPIE 1859, p.24, (1993).

6. L. W. Green, G. A. McRae, and P. A. Rochefort, “Selective resonant ionization of zirconium isotopes using intermediate-state alignment,” Phys. Rev. A**47**, 4946 (1993).
7. L. C. Balling and J. J. Wright, “Use of angular momentum selection rules for laser isotope separation”, Appl. Phys. Lett. 29, p.411 (1976)
8. G. I. Bekov, A. N. Zherikin, V. S. Letokhof, V. I. Mishin and V. N.Fedseev; JETP Lett. 33, p.450, (1981).
9. E. LeGuyadec, J. Ravoire, R. Botter, F. Lambert and A. Petit, “Effect of a magnetic field on the resonant multistep selective photoionization of gadolinium isotopes”, Opt. Commun. 76, p.34, (1990).
10. S. Villani, *Uranium Enrichment*, Topics in Applied Physics, Vol. 35 (Springer-Verlag, Berlin, Hidelberg, New York, 1977).
11. A. Corney, *Atomic and Laser Spectroscopy* (Clarendon, Oxford ,1977) Chap. 15.
12. W. Gawlik, D. Gawlik and H. Walter, *The Hanle Effect and Level-Crossing Spectroscopy*, eds. G. Moruzzi and F. Strumia (Plenum, New York,1991) Chap.2.
13. A. Messiah, *Mecanique Quantique* (Dunod, Paris, 1959).Translation: Messiah Ryoshi Rikigaku (Tokyo Tosho, Tokyo, 1971), Chap.13 [in Japanese].
14. H. Niki, I. Kitazima and Y. Izawa, “Magnetic field effect on laser isotope separation based on polarization selection rules”, Jpn. J. Appl. Phys. Vol. 37, pp.3343-3347, (1998).
15. B. W. Shore, *The Theory of Coherent Atomic Excitation* (John Wiley & Sons, 1990).
16. M. P. Sharma and J. A. Roversi, Phys. Rev. A29, p.3264, (1984).