

ORIGINAL PAPER

Selectivity and Efficiency of Laser Isotope Separation Processes of Gadolinium

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Isotopic selectivity and ionization efficiency are discussed in the isotope separation processes of Gd based on polarization selection rules and based on isotope shift. Enrichment of ¹⁵⁷Gd to 90% was experimentally demonstrated using both methods together. The features of the absorption spectra of Gd in yellow-red and blue regions were summarized. Absorption spectra that are suitable for separation process using isotope shift were examined and an obtainable selectivity and efficiency are quantitatively calculated.

KEYWORDS: *laser isotope separation, gadolinium, polarization selection rule, isotope shift, hyperfine structure*

I. Introduction

Gadolinium (Gd) is known to have a large neutron absorption cross section and currently used in light water nuclear reactors as a burnable poison. Gadolinium has seven isotopic components. Among them the odd isotopes have very large cross sections (¹⁵⁷Gd: 254,000 b, ¹⁵⁵Gd: 61,000 b) and isotope separation of these two or ¹⁵⁷Gd is awaited for economizing on fuel consumption.

In a laser isotope separation process the target isotope is usually selectively excited by a narrow-band laser and then ionized by another laser, while the other isotopes remain in their initial state. Isotope-enriched ions are collected by an external electric field. In this method the spectral broadening is required to be narrower than the isotope shift in order to obtain a high isotopic selectivity. In the atomic vapor laser isotope separation (AVLIS) with a well collimated atomic beam and a sufficiently narrow-band laser, a high isotopic selectivity can be obtained at a small laser power. Selectivity decreases with a laser power density due to power broadening. On the other hand the ionization efficiency increases with a laser power. Selectivity and efficiency are always important in the process such as isotope separation.

Isotope separation method using polarization selection rules can be also applicable to Gd because its ground state has a small angular momentum quantum number. In this method polarized lasers are used to prepare aligned states.¹⁾ Ionization of even isotopes is prohibited by the selection rules, while hyperfine interactions in the odd isotopes allow excitation and ionization of these isotopes. This method does not require a narrow-band laser. Selectivity may be decreased when there exists an external magnetic field at the laser-atom interaction region or when the laser polarizations are imperfect. Studies on laser Gd enrichment have been per-

formed using this method so far. The method using isotope shift, however, may give a high ionization efficiency and selectivity.

In this paper we describe the isotopic selectivity and the ionization efficiency in the laser isotope separation of Gd based on polarization selection rules and based on isotope shift. Total efficiency depends also on the ratio of the produced atomic vapor irradiated by lasers and other things, which were not discussed here.

II. Absorption Spectra

When an atomic vapor of Gd is generated by heating a metallic Gd, the ground state and the four metastable states are initially populated. Therefore these states are chosen to be the initial states in the atomic vapor isotope separation process of Gd. Among seven isotopic components ¹⁵⁷Gd and ¹⁵⁵Gd have a nonzero nuclear spin ($I=3/2$) and have hyperfine structures in their energy state. Absorption spectra were measured in the wavelength region of 560–600 nm²⁾ using a single-mode ring dye laser. Twenty spectra showed that the widths of hyperfine spread of odd isotopes are very much different although the spectral patterns are similar for the even isotopes. The values of isotope shift per unit mass change lie in the range of 0.4–0.7 GHz depending on the transition. Two examples of schematically drawn absorption spectra are shown in **Fig. 1**. Here the relative laser frequency is shown along the transverse axis. The spectrum of 999 to 18,070 cm⁻¹ level shows the widest spectral regions of the odd isotopes (Fig. 1(a)). The spectral region of ¹⁵⁷Gd partially overlaps with that of ¹⁵⁵Gd and contains ¹⁵⁶Gd line. In the ¹⁵⁷Gd separation using isotope shift, high isotopic selectivity and excitation efficiency cannot be achieved simultaneously in this case. On the other hand the spectrum of 533 to 17,974 cm⁻¹ level shows the narrowest hyperfine spreads (Fig. 1(b)). There is no spectral overlap between isotopes. This type of spectrum is desirable for laser isotope separation using isotope shift.

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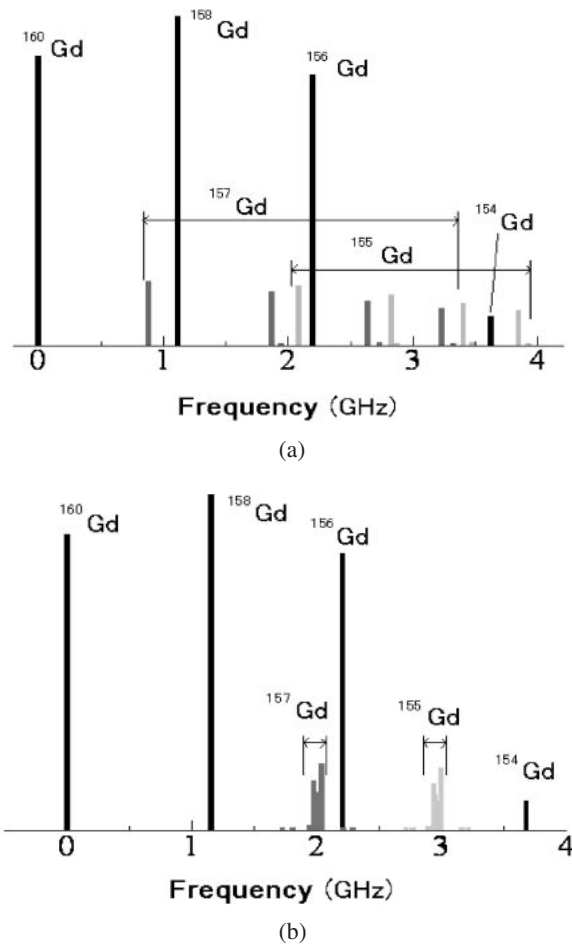


Fig. 1 Gd absorption spectra for (a) the 999–18,070 cm^{-1} transition and for (b) the 533–17,974 cm^{-1} transition

Considering the ionization potential of Gd (6.14 eV), spectral data in the above-mentioned yellow-red wavelength region are useful in the three-step photoionization process. Recent developments in semiconductor laser provide narrow-band tunable light sources of good controllability in the blue and ultraviolet region. Light source in this wavelength region may realize the two-step photoionization process. Recently Gd absorption spectra have been measured in the 400 nm region using an extra-cavity GaN laser.³⁾ The spectral patterns are similar to those in the yellow-red region. The value of isotope shift per unit mass change varies in the range of 0.6–1.0 GHz in this wavelength range. A very narrow hyperfine spread can be found in the spectrum of 999 to 25,661 cm^{-1} level. Isotopic selectivity and ionization efficiency in a separation process employing this type of spectrum will be discussed later.

III. Isotope Separation Based on Polarization Selection Rules

Researches on laser isotope separation of Gd based on polarization selection rules have been reported so far.^{4–8)} Basic principle of this method is shown in **Fig. 2**. In this method the ground state with $J=2$ angular momentum is chosen to be the initial state for three-step photoionization and the final

state should be an autoionizing state with $J=0$. When three resonant lasers with linear polarization in the same direction are used to excite Gd atoms through a pathway with $J=2 \rightarrow 2 \rightarrow 1 \rightarrow 0$ or $J=2 \rightarrow 1 \rightarrow 1 \rightarrow 0$, the odd isotopes with $I=3/2$ nuclear spin are selectively ionized as shown in **Fig. 2**. Ionization of even isotopes is prohibited in this case. Circularly polarized lasers can be used as well when they are polarized in the same direction.

Here we show two experimental results of isotope separation of Gd. The ground state ($J=2$, 0 cm^{-1}) was the initial state. The energy states at 17,381 cm^{-1} ($J=2$) and 34,587 cm^{-1} ($J=1$) were chosen to be the first and the second excited states, respectively, and the autoionizing state at 50,625 cm^{-1} was the final state. Three dye lasers pumped by a frequency-doubled Nd:YAG laser were used as the excitation source. The laser pulses were 5 ns in duration. Temporal delay between the dye laser pulses were adjusted by changing the optical path length. A quartz $\lambda/4$ plate and a polarizing filter (extinction ratio: $>500:1$) were used to obtain the linear polarization. Three dye laser beams were introduced in a vacuum chamber and irradiated the Gd atomic beam generated by an electron-beam heating of metallic Gd. The ions generated at the interaction region were deflected by an electric field and their mass spectrum was analyzed by a 0.9 m time-of-flight mass spectrometer. The dye lasers were operated at multimode and the bandwidths were 7 GHz (FWHM). **Figure 3** shows the schematic absorption spectrum of the first-step excitation. All the spectral lines lie in the range of 4 GHz, that is narrower than the laser bandwidth. An example of the mass spectrum is shown in **Fig. 4**. Considering the natural abundances (^{160}Gd : 21.86%, ^{158}Gd : 24.84%, ^{157}Gd : 15.65%, ^{156}Gd : 20.47%, ^{155}Gd : 14.80%, ^{154}Gd : 2.18%, ^{152}Gd : 0.20%), it can be seen that the ionization selectivity of the odd isotopes seems to be perfect. As shown in **Fig. 3**, the spectral regions of ^{157}Gd and ^{155}Gd are spectrally separated though ^{156}Gd line lies in the spread ^{157}Gd spectral region. Therefore further enhancement of ^{157}Gd concentration by using the relatively narrow-band first excitation laser tuned to ^{157}Gd . **Figure 5** shows an experimental result when the spectral width of the first excitation laser was narrowed to approximately 500 MHz using an intra-cavity etalon. Ionization of ^{155}Gd was suppressed and the concentration of ^{157}Gd was enhanced to about 90%. Both the isotope shift between ^{157}Gd and ^{155}Gd and the polarization selection rules were successfully utilized in this case. Ionization of even isotopes, however, would increase with increasing the laser power that leads to selectivity loss, when the degree of polarization of a laser is degraded or the polarization directions of three lasers are not perfectly parallel. As shown in **Fig. 2(b)**, among 20 magnetic sublevels of the initial state 14 sublevels are optically connected to the final state. Therefore in principle 70% of ^{157}Gd atoms in the initial state can be ionized. However it may not be necessarily easy because the transition dipole moment between sublevels varies much with magnetic quantum number m_F . Further investigation is necessary to know the relationships between the ionization efficiency of the odd isotopes and the isotopic selectivity that are practically obtainable.

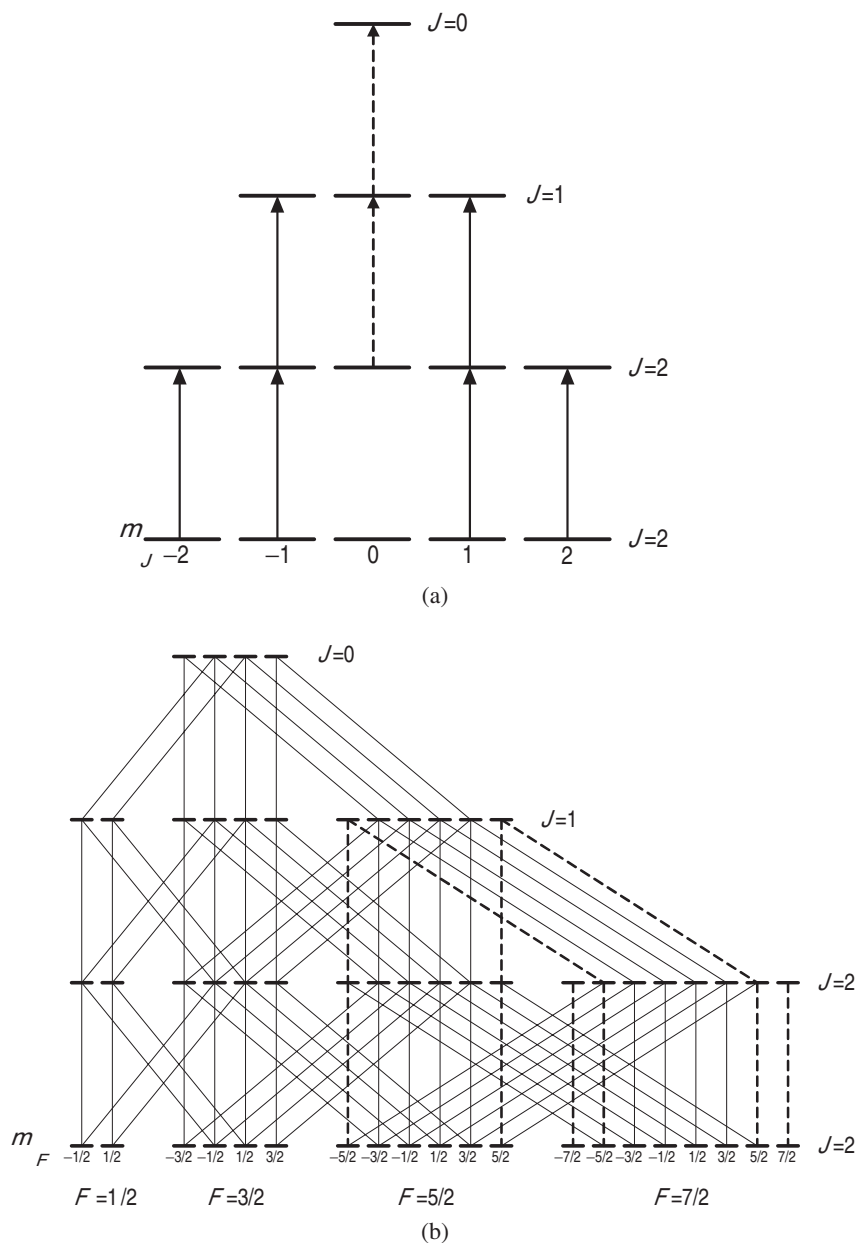


Fig. 2 Allowed transitions between magnetic sublevels of Gd through the pathway with $J=2 \rightarrow 2 \rightarrow 1 \rightarrow 0$ for (a) an even isotope and (b) an odd isotope

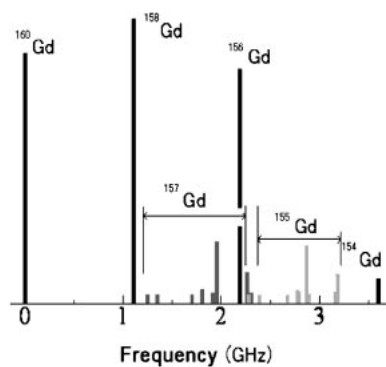


Fig. 3 Gd absorption spectrum of the $0-17,381 \text{ cm}^{-1}$ transition used as the first excitation step in the isotope separation experiment based on polarization selection rules

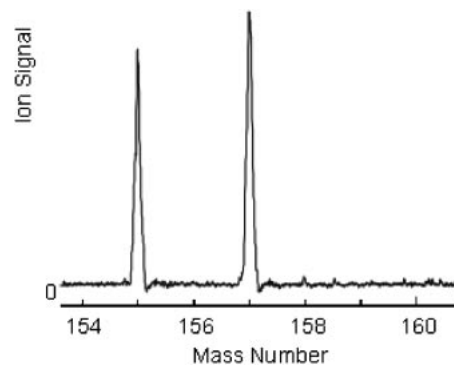


Fig. 4 Experimentally obtained mass spectrum. Ionization of the even isotopes is completely suppressed

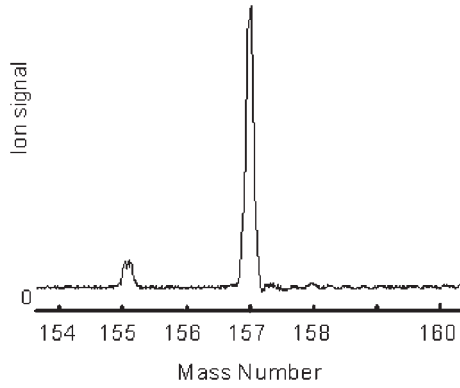


Fig. 5 Experimentally obtained mass spectrum using a 500 MHz bandwidth laser for the first-step excitation

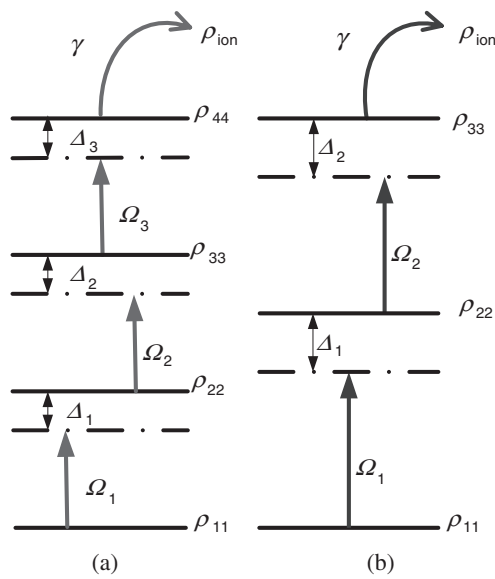


Fig. 6 Schematic diagrams of (a) three-step and (b) two-step photoionizations for numerical analyses

IV. Isotopic Selectivity and Ionization Efficiency in Separation Process Based on Isotope Shift

We show the calculated results on the ionization efficiency (the number of laser-ionized ^{157}Gd atoms/the number of ^{157}Gd atoms in the initial state before laser irradiation) and the concentration of enriched ^{157}Gd . It is assumed that ^{157}Gd with negligibly small hyperfine spread is selectively ionized based on isotope shift by two-step or three-step photoionization as shown in **Fig. 6**. Calculations were done by solving the equations describing atomic excitation dynamics written by density matrices. Only two isotopes, ^{157}Gd and ^{156}Gd , were taken into account because the spectral shifts of the other isotopes from ^{157}Gd are much larger than that of ^{156}Gd . Actually, in the $533\text{--}17,974\text{ cm}^{-1}$ transition shown in Fig. 1(b), the spectral shift of ^{158}Gd from ^{157}Gd is 4.2 times larger than that of ^{156}Gd .²⁾ In Fig. 6 Ω_i and γ denote the Rabi frequency and the autoionization rate, respectively, and Δ_i is the detuning (the difference between

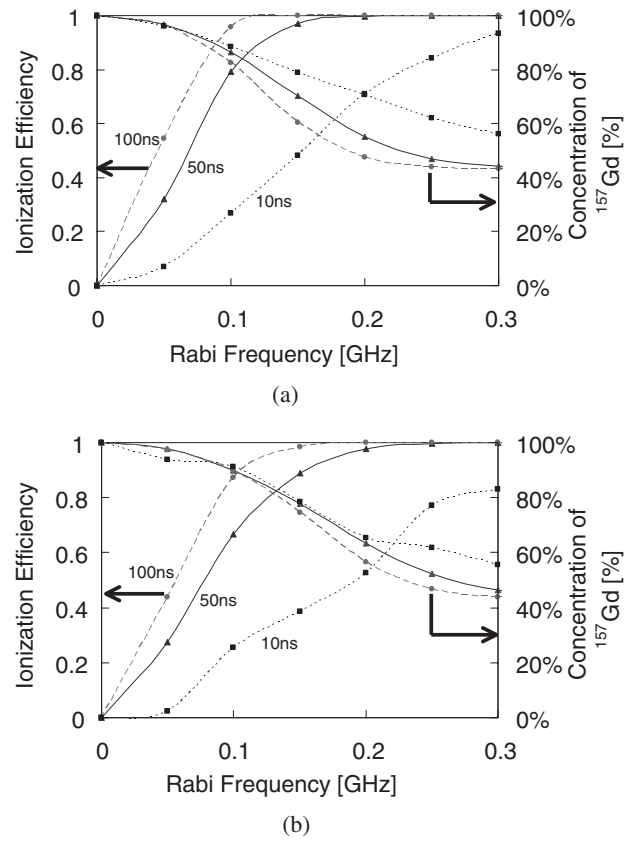


Fig. 7 Calculated ionization efficiency and concentration of ^{157}Gd as a function of Rabi frequency for laser pulse widths of 10, 50 and 100 ns

atomic resonance frequency and laser frequency). Every laser frequency is tuned to each resonance frequency of ^{157}Gd . Therefore the value of Δ_i is the isotope shift. According to the measured spectral data^{2,3)} the Δ_1 's were set to 200 and 280 MHz for the three-step and the two-step photoionization of ^{156}Gd , respectively. From the absorption measurements for the second excitation step,⁹⁾ the isotope shifts were twice or more larger than those for the first excitation step. And the hyperfine spread of ^{157}Gd was larger than the isotope shift in every spectrum. Therefore the isotopic selectivity would not be expected in this excitation step and also in the third step. Assuming that no effective selectivity is expected in the second and in the third excitation steps, the Δ_2 and Δ_3 were set to zero. The Rabi frequencies were equal for all the excitation steps and the autoionization rate was set to 1 GHz for all the calculations. Calculated ionization efficiency and concentration of ^{157}Gd defined above are shown as a function of Rabi frequency for the two-step and the three-step excitation scheme, in **Figs. 7(a) and (b)**, respectively. Here the value of Rabi frequency is proportional to the square root of laser power density. The results are shown for laser pulse widths of 10, 50 and 100 ns. Generally, as the Rabi frequency increases, the ^{157}Gd concentration decreases because the ionization of ^{156}Gd is enhanced due to power broadening. The ionization efficiency increases with the laser pulse energy. It is seen from Fig. 7(b) that both the ionization efficiency and the ^{157}Gd concentration can be

kept more than 80% simultaneously at Rabi frequencies around 0.1 GHz with the laser pulse widths of 50 ns or longer. If an effective isotope shift is found in the second or third excitation step, the selectivity would be improved.

V. Summary

In laser isotope separation of Gd, isotopic selectivity and ionization efficiency were discussed. The concentration of ^{157}Gd was successfully enhanced up to about 90% in the experiment using the polarization selection rules together with the isotope shift between ^{157}Gd and ^{155}Gd . The features of the absorption spectra of Gd in yellow-red and blue regions were summarized. Absorption spectra with very narrow hyperfine spreads that are suitable for separation process using isotope shift were examined. By solving the equations describing atomic excitation dynamics written by density matrices, the ionization efficiency and the ^{157}Gd concentration were calculated. It was shown that these two values can be kept to more than 80% at Rabi frequencies around 0.1 GHz with the laser pulse widths of 50 ns or longer.

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