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NUMERICAL EVALUATION OF THREE-STEP SELECTIVE PHOTO-IONIZATION OF GD^{157} AND GD^{155} ISOTOPES*

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Abstract – A three-step selective photo-ionization for Gd^{155} and Gd^{157} isotopes by pulsed laser was evaluated numerically. Due to large thermal neutron absorption cross sections, these two isotopes in natural gadolinium are good candidates as burnable poison to control reactivity early in the fuel cycle and allow extended fuel burn up. Rate equations have been used to calculate the dynamics of the isotope populations, ionization rate, and selection factor. Doppler broadening, as well as power broadening for atomic energy levels, has been considered in the calculations. The effects of laser intensity, frequency bandwidth, and laser detuning in the ionization process have also been assessed. By applying a variety of conditions, the sensitivity of different parameters in the ionization process was investigated. The results show that the ionization yield increases with the intensities of lasers, and becomes saturated at some specific values. Depending on the energy bandwidth of each state, laser bandwidth and the detuning of lasers greater than particular values can affect the ionization yield. Due to small and finite isotope shifts of energy states, the selection factor can be influenced by detuning and laser bandwidth. From the results, it can be inferred that for Gd^{155} and Gd^{157} isotopes, ionization is more sensitive to the first and second laser bandwidth and detuning. From these results, optimum selection factors and ionization by using appropriate laser intensities, bandwidth, and detuning can be achieved.

Keywords – Photo-ionization, multi-step photo-ionization, selective photo-ionization, pulse laser, photon-atom interaction

1. INTRODUCTION

Atomic photo-excitation and ionization by pulsed lasers have been of interest for many years [1-5]. The laser photo-ionization is not only a very useful technique for the investigation of the properties of laser matter interaction, but also for several important applications [6-8]. In the process of the laser photo-ionization, the resonance effect plays a very important role in the ion yield and selectivity.

Some experiments have been performed on the resonant and near-resonant effects in the multi-photon ionization process [9-13]. The development of methods of stepwise excitation of high-lying states of atoms by radiation of pulsed tunable dye lasers has made it possible to investigate auto-ionization states. Auto-ionization states are of scientific interest for the understanding, identification, and interpretation of atomic spectra of multi-electron atoms.

The search for finding methods in separating chemical elements and isotopes have also been simulated by the widespread use of various isotopes from ultra pure materials in power plants, industry, and scientific research. The laser method for separating isotopes and fabricating ultra pure

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materials in which laser light interacts resonantly with the material, has been a good choice among the others due to less energy consumption and cost. However, stepwise photo-excitation, and the ionization technique are expensive processes and a theoretical evaluation can reduce the investigation costs.

Natural gadolinium is a good burnable poison to control reactivity early in the fuel cycle and allow extended fuel burn up. The active isotopes in this control are the odd isotopes (Gd^{155}, and Gd^{157}) which have the largest thermal neutron absorption cross sections relative to the other isotopes. The thermal neutron absorption cross section for Gd^{155} and Gd^{157} are 61000 and 255000 Barns, and their abundance are 14.8%, and 15.65%, respectively.

In this paper we have applied rate equations to evaluate Gd^{155} and Gd^{157} isotopes in a three step selective photo-ionization process, taking into account some conditions for both light source, namely laser, and the atomic source. Under a particular condition, the population of all transition levels as well as the ionization level can be described by the rate equations. This condition can be satisfied when the stimulated rates of R_i dominate the various decay rates T_i, i.e. $R_i > 1/T_i$ [14]. This will be discussed in more detail in the next section. The effects of laser bandwidth, together with the Doppler broadening due to atom velocity distribution, as well as power broadening in the selection factor and ion density were inspected. The isotope shift for the states of Gd^{155} and Gd^{157} were also taken into account, and the detuning effects by mismatching the laser frequency from the atomic transition frequency for all levels were examined.

2. THEORY AND THE CALCULATION METHOD

For the study of multi-step excitation and ionization, two kinds of theoretical treatment have been introduced so far in some literature, namely classical and quantum mechanical methods. Ackerhalt et al. [14], and Shore, and Johnson [15] evaluated the two methods and their limitations very well. The most complicated equation, generally approved to provide the most accurate description, would be a density-matrix equation of motion (a generalized Bloch equation) treating both phase preserving (coherence) processes and phase destroying (relaxation) processes. When incoherent relaxation processes are unimportant (stimulated processes dominate spontaneous emission, collisions are negligible, and lasers are coherent) then the complexity of the problem can be greatly simplified by replacing the Bloch equation with the Schrodinger equation. On the other hand, when phase relationships are unimportant and relaxation dominates coherence, the Bloch equation is reduced to an equation for populations-rate equation (PRE). However, it is shown by Ackerhalt et al. [14] that under some circumstances, the rate equation can be used to study multi-step excitation, even if the exciting lasers have very narrow bandwidths, high intensities, and are tuned to resonance with the numerous transitions in the ladder.

The PRE's do not provide a complete description of any excitation process. Their structure is suitable only for processes in which the populations change monotonically. It is well known that non-monotonic pulsations of level populations can occur if an atom is subjected to intense coherent nearly resonant radiation. These population pulsations occur at the Rabi rate Ω, given by [16]:

$$\Omega = \left[(2dE/\hbar)^2 + (\omega_L - \omega_A)^2\right]^{1/2} \tag{1}$$

Where d is the transition dipole matrix element, E is the amplitude of the laser field, ω_L and ω_A are the laser and atomic frequency, respectively.

It is shown [17] that under the condition of $\Omega > \Gamma$ where Γ is the transition line-width, the PRE's are no longer adequate to describe the excitation process. That is, one has smooth monotonic population flow through the levels of the atom (and valid PRE's) only when $\Omega < \Gamma$. On the other hand, multi-photon transitions leading to ionization or dissociation are effectively irreversible at the final step. Therefore, the last transition can be stimulated just as strongly without producing Rabi cycling in the transition. However, it is also shown [14] that under the condition of:

$$R_4 > R_3 > R_2 > R_1 \tag{2}$$

Where R_i is the stimulated rate for step *i*, the rate equation is sufficient to describe the population of all the lower transitions as well, so long as the laser powers are high enough. This condition can be satisfied when the stimulated rates of R_i dominate the various decay rates $1/T_i$, i.e. $R_i > 1/T_i$.

The purpose of this paper is to analyze and optimize a three-stage scheme for selective photo-ionization of Gd^{155} and Gd^{157} isotopes using PRE's under non-stationary conditions. We allow non-uniform broadening for the resonant atomic absorption lines and consider isotope shift for all energy levels, to take into account possible loss of selectivity caused by overlap of the absorption lines of different isotopes. Using appropriate laser polarization, the even isotopes of Gd can be eliminated in a selective photo-ionization process [18]. In order to formulate the problem, we considered an atomic beam of known density consisting of a mixture of isotopes "a", and "b". The density distribution of the atomic beam with respect to the projection of the velocities on the axis is supposed to be known. Three pulsed laser beams in a direction parallel to the x-axis interact on the atomic beam that is shown schematically in Fig. (1-a). The atoms irradiated by such pulses have energy states that are shown in Fig. (1-b). The interaction of the laser pulses with the atoms can lead them to their auto-ionization states. The pulses, with equal duration, are assumed to be Gaussian in time and frequency domain as a more realistic condition in experiments. The intensity of each laser pulse can be given by:

$$I(\omega,t) = \frac{2\sqrt{\ln 2/\pi}}{\Delta\omega_L} I(t) \exp\left\{-\left[2\sqrt{\ln 2}\,\frac{(\omega-\omega_L)}{\Delta\omega_L}\right]^2\right\} \tag{3}$$

In which ω_L and $\Delta\omega_L$ are the laser frequency at maximum intensity and its FWHM respectively. $I(t)$ is the time varying part of the intensity for each laser pulse, and is given by:

$$I(t) = I_0 \exp\left\{-\left[2\sqrt{\ln 2}\,\frac{(t-t_0)}{\tau_p}\right]^2\right\} \tag{4}$$

where I_0 and τ_p are the maximum intensity and the pulse duration of the laser, respectively. In relation (4), t_0 is the time when the maximum laser intensity occurs in the time domain. We also consider the case in which the laser pulse length is much shorter than the time required for the atoms to cross the beam.

The densities of the energy levels i, $n_i(v_z,t)$ are described by rate equation as follows:

$$\frac{dn_1}{dt} = w_1\left(n_2/g - n_1\right) + A_{21}n_2 \tag{5}$$

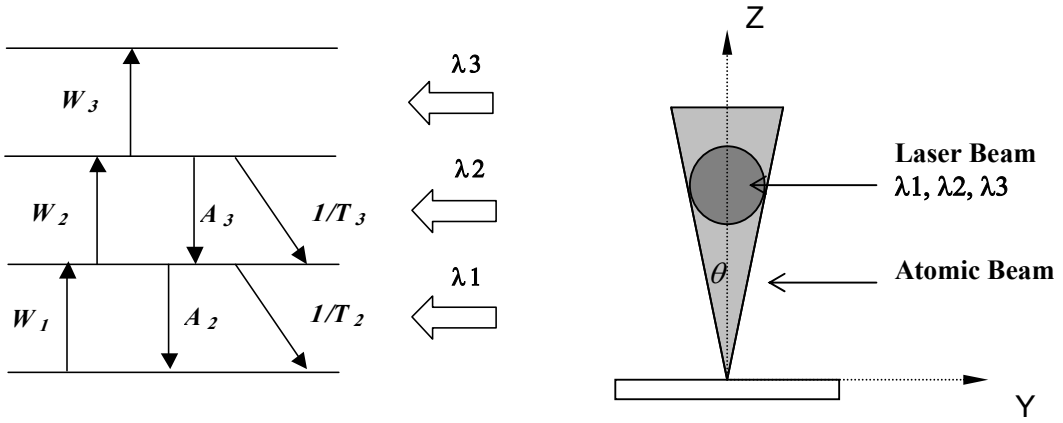

Fig. 1. Schematic diagram of a three step, four level photo ionization processes

$$\frac{dn_2}{dt} = w_1\left(n_1 - n_2 / g\right) - w_2\left(n_2 - n_3 / g'\right) - \frac{1}{T_2} n_2 + A_{32} n_3 \tag{6}$$

$$\frac{dn_3}{dt} = w_2\left(n_2 - n_3 / g'\right) - w_3 n_3 - \frac{1}{T_3} n_3 \tag{7}$$

$$\frac{dn_{4m}^{+}}{dt} = w_3 n_3 \tag{8}$$

Where T_2, T_3 are the lifetimes of levels 2 and 3, respectively, n_{4m}^{+} is the ion density of isotope "*m*", and A_{ul} is the appropriate Einstein coefficient for the transition $u \to l$. The factors g, g' are defined by:

$$g = \frac{g_2}{g_1} \tag{9}$$

$$g' = \frac{g_3}{g_2} \tag{10}$$

Where g_i is the degeneracy of the energy level *i*.

In relations (5)-(8), $w_i\left(v_z, t\right)$ is given by:

$$w_i\left(v_z, t\right) = \int_{-\infty}^{+\infty} \frac{4\pi\sigma_i\left(\omega, v_z\right) I_i\left(\omega, t\right)}{h\omega_0} d\omega \tag{11}$$

Where h is the plank constant, ω_0 is the central frequency of the transition, and $\sigma_i\left(\omega, v_z\right)$ is the homogeneous absorption cross section of the step i at frequency ω, and for atoms with velocity v_z, and is given by [19]:

$$\sigma_i(\omega, v_z) = \frac{\lambda_0^2}{4\pi}\frac{g_u}{g_l}\frac{A_{ul}\Gamma_{lu}}{\left[\omega - \left(1 - \frac{v_z}{c}\right)\omega_0\right]^2 + \Gamma_{lu}^2} \tag{12}$$

In the above formula, λ_0 is the resonance central wavelength of step i, A_{ul} is the Einstein coefficient for the transition $u \to l$, and g_u / g_l is the degeneracy ratio of u and l states, and Γ_{lu} is the FWHM of the homogeneous transition including power broadening and is given by:

$$\Gamma_{lu} = \frac{1}{2}\left[\sum_p \gamma_{pl} + \sum_p \gamma_{pu}\right] \tag{13}$$

In which γ_{pl} and γ_{pu} are the rate of all coherent and non-coherent transitions $p \to l$ and $p \to u$. For transition $1 \to 2$, for example, Γ_{lu} can be given by:

$$\Gamma_{12} = \frac{1}{2}\left[w_1 + \frac{g_1}{g_2}w_1 + w_2 + \frac{1}{T_2}\right] \tag{14}$$

Where T_2 is the lifetime of the second state.

Assuming an atomic beam with divergence ϑ, and a velocity distribution function $g(v_x)$ in the laser beam direction, the average density of level i can be given by:

$$n_i(t) = \int_{-\infty}^{+\infty} n_i(v_z, t) g(v_z) dv_x \tag{15}$$

Where $n_i(v_z, t)$ calculated from relations (5)-(8) and $g(v_z)$ is given by [19]:

$$g(v_z) = \frac{1}{\sqrt{\pi} v_P \sin\vartheta} \exp\left(-\left[\frac{v_z}{v_P \sin\vartheta}\right]^2\right) \tag{16}$$

In relation (16), v_P is the most probable velocity and is related to the Doppler bandwidth of the atomic beam $\delta\upsilon_D$ by:

$$v_P = \frac{\pi}{\sqrt{\ln 2}}\frac{c\delta\upsilon_D}{\omega_0} \tag{17}$$

Where ω_0 is the transition central frequency, and c is the speed of light in vacuum. If one solves relations (3)-(17) simultaneously for isotopes "*a*", and "*b*", the ion density of the two isotopes, n_{4a}^+ and n_{4b}^+, the ionization coefficient for isotope "*a*" or "*b*", $\alpha = n_4^+ / n_4$, and selection factor $\delta = n_{4a}^+ / n_{4b}^+$ can be calculated.

3. RESULTS AND DISCUSSION

In order to verify the validation of the model explained in the previous section, relations (3-17) were solved simultaneously for the Gd atom with energy levels, and transitions employed by Bekov [20]. All required atomic and laser parameters that were used in the calculation summarized in Table 1.

Figure 2 shows the ion yield versus third laser intensity. The saturated intensity for the third laser from Ref. [20] is about $0.8\times10^{-3}\,J/cm^2$. From Fig. 2, a value of $0.9\times10^{-3}\,J/cm^2$ can be found for the saturated intensity of the third laser. It can be seen that the calculated saturated intensity by our model for the third laser is in good agreement with that of the experiment reported in Ref. [20]. It must be noted that the measured ion yield in Ref. [20] is expressed in unit (mV), while it is described in a relative unit in Fig. 2.

Table 1. Laser and Gd spectroscopic parameters used for verification of the model (From Ref. [20])

$\lambda_1(\overset{o}{A})$	$\lambda_2(\overset{o}{A})$	$\lambda_3(\overset{o}{A})$	$I_1(\mu J/cm^2)$	$I_2(\mu J/cm^2)$	$\delta\upsilon_D(GHz)$	$\delta\upsilon_L(GHz)$	$\tau_P(n\,sec)$
5618	6351.7	6133.5	250	50	2.1	0.9	7

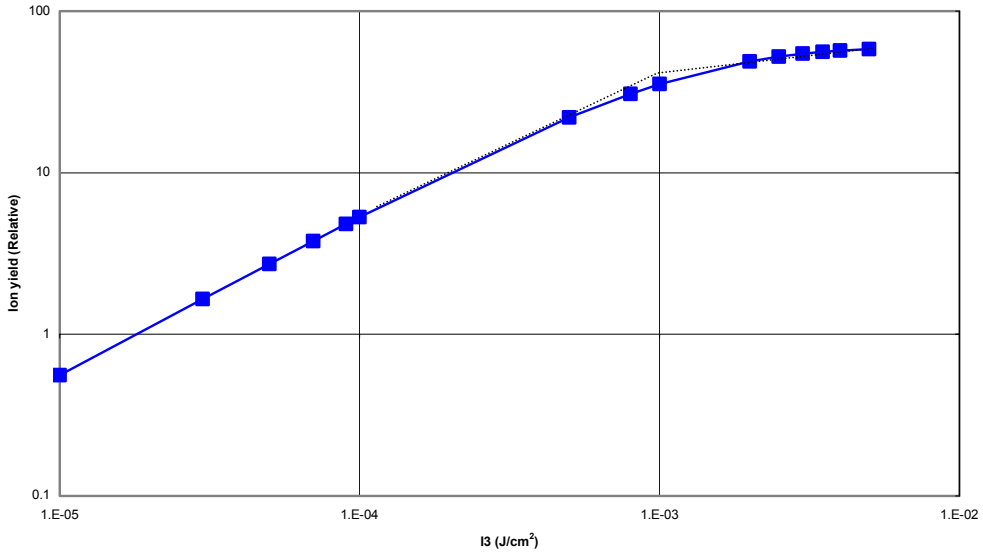

Fig. 2. The ion yield versus third laser intensity using the experimental condition of Ref. [20]

Applying the spectroscopic parameters of isotopes Gd^{155} and Gd^{157} [18], the effects of laser bandwidth, detuning, intensities on ionization yield, and selection factor were investigated. The spectroscopic parameters of isotopes Gd^{155} and Gd^{157} are summarized in Table II. We assumed a bandwidth of 230 MHz, and a duration of $45n\,sec$ for the lasers as employed by Ref. [18]. For the atomic beam the divergence was 10 degrees, and Doppler bandwidth $\delta\upsilon_D$ was assumed to be $250\,MHz$. The ionization coefficient and selection factor were calculated for different laser intensities, assuming the three lasers to be tuned on Gd^{157} energy states. Figures 3 and 4 show the ionization coefficient of Gd^{157} and Gd^{155} against the first and second laser intensities, and for the different intensity of the third laser. For both isotopes, the ionization coefficient rises with the intensity of the first and second laser, but reaches a saturated value at some specific intensity. These values are nearly 0.1, 1.1, and 2.5 mJ/cm^2 for the first to the third laser, respectively, for the conditions of our calculations. With an intensity of 2.1 mJ/cm^2 for the third laser, optimum values of 0.55, and 0.003 were obtained for the ionization coefficient of isotopes Gd^{157} and Gd^{155} respectively. From the relations it is obvious that the ionization yield saturation is mostly due to the finite lifetime of states, and therefore limitation of the interaction at the higher intensities, but the

power broadening of the lasers on the energy bandwidth are also effective. The saturation intensity for each laser is different. This can be explained by the fact that the broadening of each state Γ_{lu} depends on w_i, the laser intensities, which are different for each state. The selection factor somehow has a different behavior. Figure 5 shows the selection factor against the first and second laser intensities, and for the different intensity of the third laser. It can be seen that the higher intensities give the smaller selection factors. For the higher intensity, the power broadening increases the interaction efficiency of Gd^{155}, as well as Gd^{157}, leading to a rise in the total ion yield, but reducing the selection factor.

Table 2. Spectroscopic parameters for gadolinium process (From Ref. [18])

$\bar{\lambda}(cm^{-1})$	J	*Lifetime* $\tau(ns)$	*Oscillator Strength,* $f(10^{-3})$	*Transition isotope shifts relative to* Gd^{157} *(GHz)*
0	2			
16923.3	2	600 ± 200	0.7 ± 0.1 for $0 \rightarrow 16923.3$	0.045 ± 0.01 for $0 \rightarrow 16923.3$
32660.8	1	800 ± 200	0.063 ± 0.01 for $16923.3 \rightarrow 32660.8$	1.571 ± 0.02 for $16923.3 \rightarrow 32660.8$
50624.7	0	0.035 ± 0.004	0.21 ± 0.3 for $32660.8 \rightarrow 50624.7$	≈ 4.5 for $32660.8 \rightarrow 50624.7$

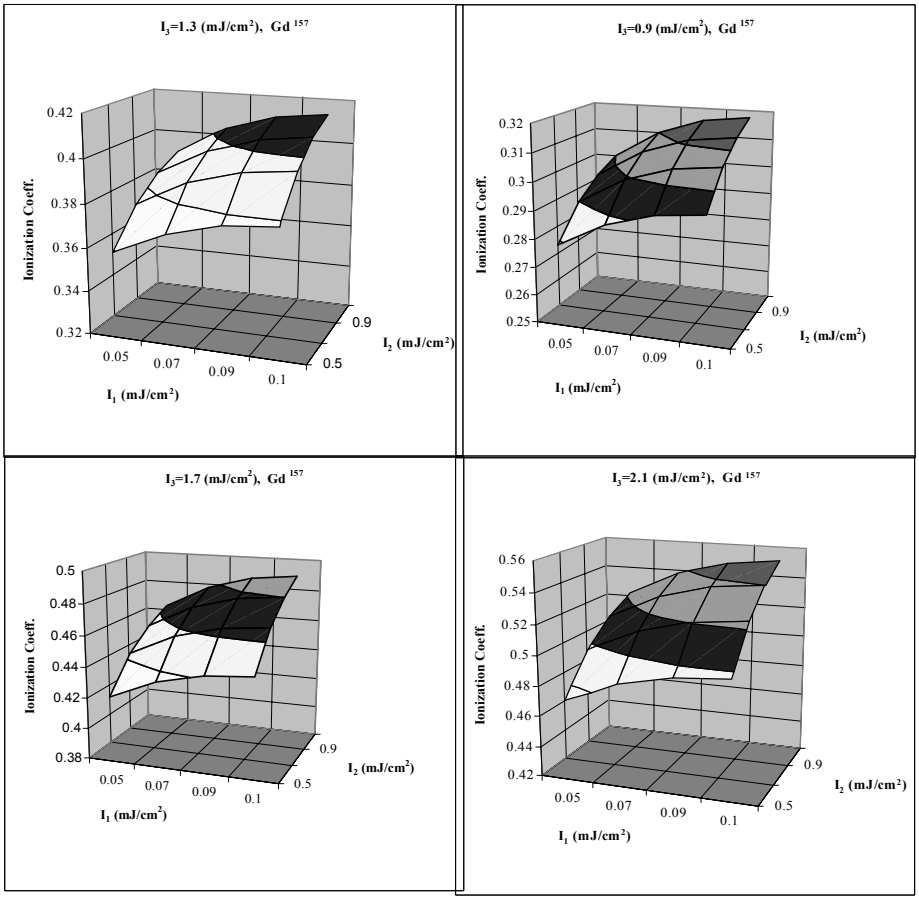

Fig. 3. The ionization coefficient of Gd^{157} against the first and second laser intensities (I_1,I_2) and for different intensity of the third laser (I_3)

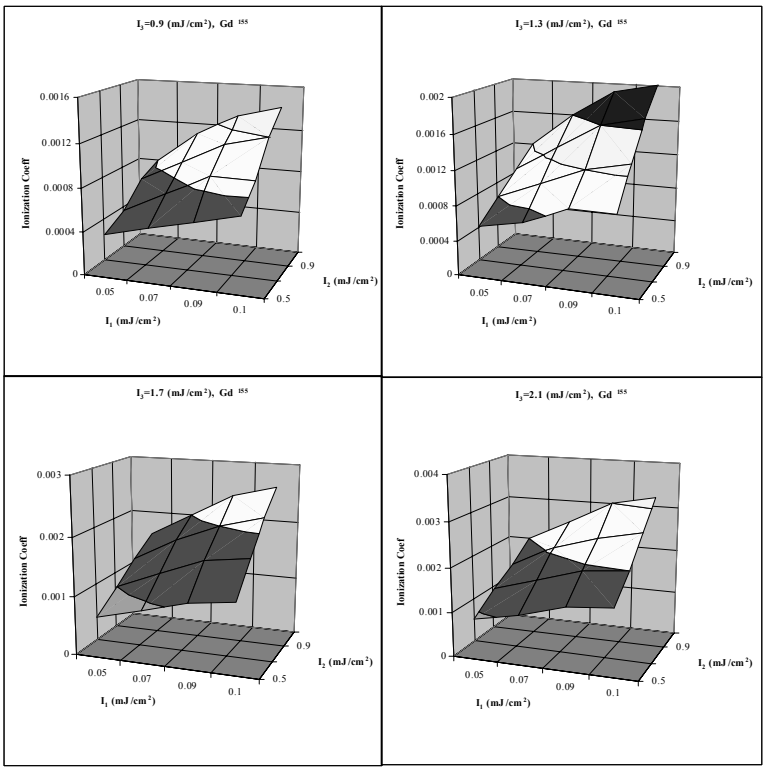

Fig. 4. The ionization coefficient of Gd^{155} against the first and second laser intensities (I_1, I_2) and for different intensity of the third laser (I_3)

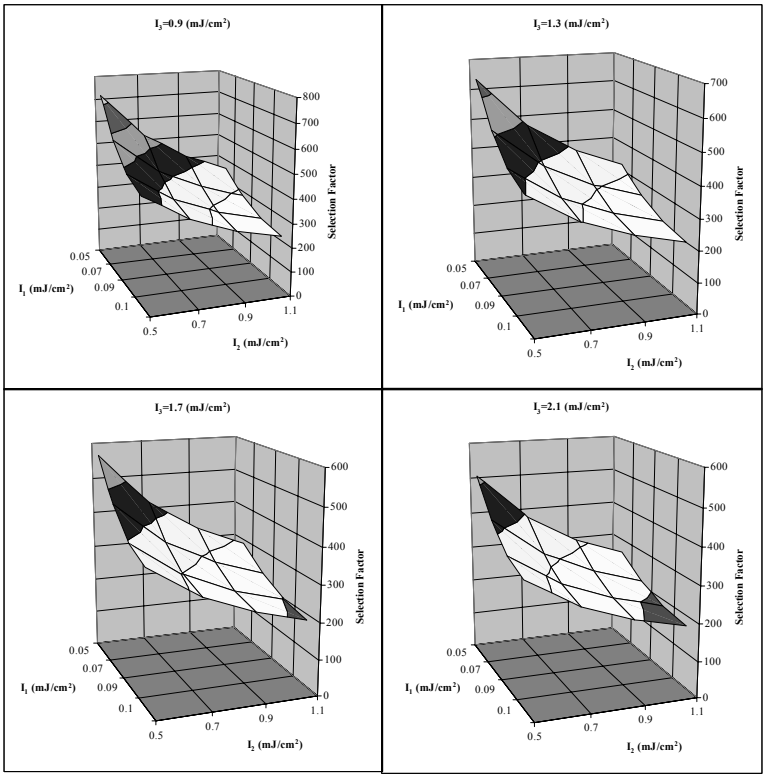

Fig. 5. The selection factor against the first and second laser intensities

Using the optimized intensities, i.e. 0.1, 1.1, and 2.5 mJ/cm^2 for the first, second, and the third laser respectively, the effects of detuning and laser bandwidth were also examined. The ionization coefficient of the two isotopes versus the first and second laser bandwidth and detuning is shown in Fig. 6. It can be seen that the ionization coefficient for Gd^{157} is not very sensitive to the detuning of the lasers to some particular values. These values are around 80, 300, and 1000 MHz for the first to the third lasers. Detuning greater than these values dramatically decreases the ionization coefficient. This is due to the influence of laser intensities, and the state lifetime on the effective atomic bandwidth Γ_{lu}. According to relation (13), Γ_{lu} is proportional to laser intensities w_i, as well as energy lifetimes $1/T_i$. For instance Γ_{23} and Γ_{34} are both proportional to w_2, and w_3, while Γ_{12} increases with w_1 and w_2. For Gd^{157}, the condition of $\Gamma_{12} < \Gamma_{23} < \Gamma_{34}$ is established and under such circumstances, the interaction will be more sensitive to the detuning of the first laser. Accordingly, the third laser can be detuned up to $1\,GHz$, since $\Gamma_{34} > 4.5\ GHz$. The interaction occurs efficiently in a more broadened range for the third laser, since both the intensity and the oscillator strength for the third laser is higher than that of the others. On the other hand, the ionization yield of Gd^{155} and the selection factor variation depends on the isotope shift. The isotope shift for the first state is small (~ $450\,MHz$), and rises to $4.5\,GHz$ for the third level. Due to a smaller isotope shift of the first state in comparison to the other two, the selection factor can be changed dramatically by small detuning of the first laser, while the second and third laser affect the ionization at larger detuning. For the third laser the ionization can occur in a broader range, since the isotope shift for this level is nearly $4.5\,GHz$.

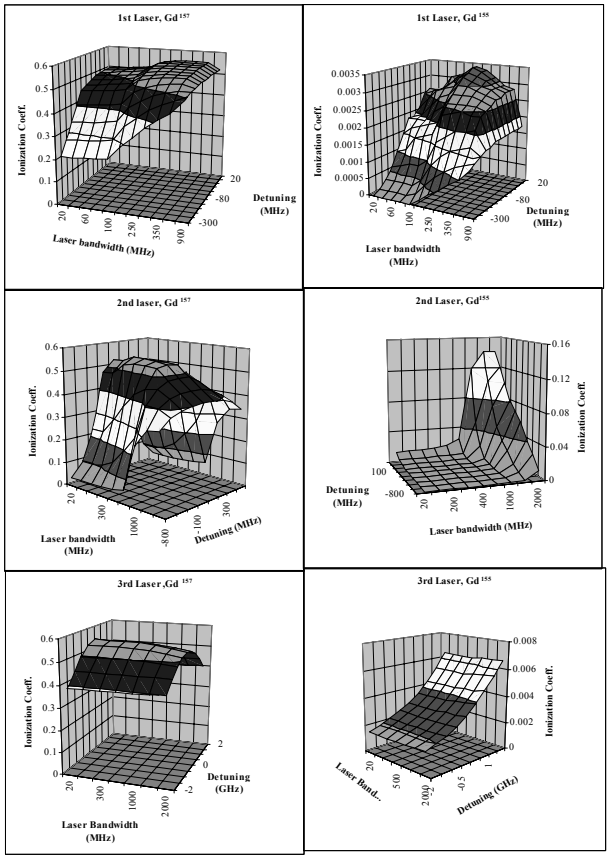

Fig. 6. The ionization coefficient of Gd^{157} and Gd^{155} verses the first and second laser bandwidth and detuning

In the next attempt we evaluated the ionization process for lasers with fixed intensities, but variable bandwidth. The first laser bandwidth up to 400 MHz does not influence the ionization significantly. This value rises to 900 MHz for the second laser. For the third laser, increasing the bandwidth to even 2000 MHz does not change the ionization extensively. These results can be interpreted by the fact that the exciting laser with a bandwidth wider than the atomic bandwidth interacts non-efficiently with the atoms due to less overlapping of the atomic and laser line shapes. Since the isotope shifts of the first and second levels are nearly 450 MHz and 1.5 GHz **,** therefore laser bandwidth greater than these values can diminish the selection factor. Obviously for the third transition, with an isotope shift of 4.5 GHz **,** a greater bandwidth is expected in order to see the change on the selection factor.

This explanation is confirmed by examining the ionization yield of Gd^{155}. Due to the very small isotope shift of the first state, similar results to that of Gd^{157} were obtained. For the second level (with isotope shift of 450 MHz), the ionization yield is nearly zero as the laser with a bandwidth of even 500 MHz will effectively ionize Gd^{157}, but any increase to the bandwidth (with little detuning) can influence Gd^{155} as well as Gd^{157}.

For the same reason, due to the finite isotope shift of the two isotopes, any broadening in atomic bandwidth increases overlapping of the two isotopes; consequently, the power broadening will diminish the selection factor.

4. CONCLUSIONS

Rate equations have been used to calculate the isotope populations, ionization rate, and selection factor of Gd^{155} and Gd^{157} in a three-step selective photo ionization process. Under the condition of our calculations, i.e. when the stimulated rates R_i dominate the various decay rates T_i, i.e. $R_i > 1/T_i$, the rate equation is able to describe the population of all the lower transitions. Doppler broadening, as well as power broadening for the atomic energy levels, has been considered in the calculations. This model was employed to describe the ionization and the selectivity of a three stepwise selective photo-ionization of Gd^{155} and Gd^{157}. The isotope Gd^{157} is a good candidate for burnable poison to control reactivity early in the fuel cycle due to its large thermal neutron absorption cross section. We have shown that there are optimized intensities for the lasers to obtain maximum selective ionization. We have also shown that the laser bandwidth and the detuning greater than some particular values can affect the selective ionization. It is believed that finite isotope shifts of each state, and the dependency of atomic bandwidth Γ_{lu} on the laser intensities, are the main basis of this behavior. The ionization field is very sensitive to the detuning and bandwidth of the first laser compared with those of the other lasers. It is concluded that for an efficient selective ionization, greater wavelength stability and tuning for the first laser is required.

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