

Selectivity Optimization in Four-Step Pulsed Photo-Ionization

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We study the selectivity and ion yield of four level atoms subject to resonant radiation from three powerful laser fields. The optimal conditions for achieving high selectivity without a significant decrease of the ion yield of the selected isotope are determined analytically and the results are compared with numerical calculations for the case of uranium. The limitations imposed by Doppler broadening due to a Maxwellian velocity distribution are also studied.

I. Introduction

A four step pulsed photo-ionization scheme is usually employed in laser isotope separation of heavy atoms [1]. Under typical conditions, Doppler broadening is much greater than the natural widths and resonant radiation of low intensity ionizes only a small fraction of the atoms. This problem is overcome by using high intensity laser radiation. Under this condition, a large fraction of undesired isotopes is ionized if the laser radiation is tuned to the center of the line for the atomic transitions of the selected isotope. Then, the selectivity drops with increasing laser intensities. This problem can be avoided with a detuning from intermediate resonance of the laser frequencies.

In reference [2], rough estimates are made for the selectivity in multi-step pulsed photo-ionization spectroscopy considering each step independent of the others. However, when power broadening becomes significant and the laser pulses act simultaneously these estimations are no longer valid and one must solve a set of coupled equations (the Schrodinger equation for the density matrix). The restriction on the selectivity becomes more severe when one needs to obtain also high ion yield.

Despite the fact that the dynamics of two and three-

step photo-ionization was studied, including the selectivity problem, in refs. [3-11], the problem of optimizing the selectivity without a significant decrease in the ionization yield was first studied in reference [12]. There, the selectivity of a three step process was studied within a certain approximation (the relaxation mode approximation) that permits to obtain analytical results. Unfortunately, a three step process is inadequate for heavy atoms if one is interested in an industrial application.

The four step process has been widely studied numerically [13-15] from the point of view of ion yield. However, selectivity considerations have not been taken into account.

We extended the relaxation mode approximation to an n-step process in [16]. Here, we apply the results of reference [16] to describe the dynamics of four step pulsed photo-ionization of atoms.

In a three level system [12], the simultaneous variation of the laser frequencies at the first and second steps $\omega_1 = \omega_{12} + \delta$ and $\omega_2 = \omega_{23} - \delta$ (ω_i are the laser frequencies and ω_{ij} are the frequencies of the transitions of the selected isotope, δ being the detuning from resonance) over a wide range of δ increases drastically the selectivity without affecting significantly the ionization probability of the selected isotope. We will show that

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in a four level system, in the limit of large detuning, the ionization probability of the selected isotope has a local maximum whenever $\omega_1 + \omega_2 + \omega_3 = \omega_{12} + \omega_{23} + \omega_{34}$. This fact leaves us with two degrees of freedom $\delta_1 = \omega_1 - \omega_{12}$ and $\delta_2 = \omega_2 - \omega_{23}$ (we consider fixed the laser intensities).

We will find optimal values for δ_1 and δ_2 in the short pulse case, with allowance for Doppler broadening. Finally we will also show results of numerical simulations. The parameters of the computation do not corresponds exactly to any atom, although their values are close to the case of uranium under ionization through a Rydberg state.

II. Relaxation mode approximation in four-step pulsed photo-ionization

We will consider the dynamics of a four-level atom subject to resonant laser radiation of frequencies ω_1 , ω_2 and ω_3 close to the atomic transitions ω_{12} , ω_{23} and ω_{34} respectively. We will assume that the frequencies ω_{ij} are not too close to each other, so that each pulse excites only its own transition. The equations for the slow density matrix elements ρ_{ij} are well described by the rotating-wave approximation [17] and can be written in the following form,

$$i \frac{dX}{dt} = (H_0 + i\Gamma)X \quad (1)$$

where X is a column vector with elements $\rho_{11}, \dots, \rho_{44}$, $\rho_{12}, \rho_{21}, \dots, \rho_{34}, \rho_{43}$, the matrix Γ depends on the decay rates and the elements of the symmetrical matrix H_0 are given by,

$$\begin{aligned} -H_{0_{1,5}} &= H_{0_{1,6}} = H_{0_{2,5}} = -H_{0_{2,6}} = v_1 \\ -H_{0_{2,12}} &= H_{0_{2,13}} = H_{0_{3,12}} = -H_{0_{3,13}} = v_2 \\ -H_{0_{3,15}} &= H_{0_{3,16}} = H_{0_{4,15}} = -H_{0_{4,16}} = v_3 \\ -H_{0_{5,5}} &= H_{0_{6,6}} = f_2 = \delta_1 \\ -H_{0_{7,7}} &= H_{0_{8,8}} = f_3 = \delta_1 + \delta_2 \\ -H_{0_{9,9}} &= H_{0_{10,10}} = f_4 = \delta_1 + \delta_2 + \delta_3 \\ -H_{0_{11,11}} &= H_{0_{12,12}} = f_2 - f_3 \\ -H_{0_{13,13}} &= H_{0_{14,14}} = f_2 - f_4 \\ -H_{0_{15,15}} &= H_{0_{16,16}} = f_3 - f_4 \\ -H_{0_{8,12}} &= H_{0_{7,11}} = H_{0_{9,13}} = -H_{0_{10,14}} = v_1 \\ -H_{0_{5,7}} &= H_{0_{6,8}} = H_{0_{13,15}} = -H_{0_{14,16}} = v_2 \end{aligned} \quad (2)$$

$$-H_{0_{7,9}} = H_{0_{8,10}} = H_{0_{12,14}} = -H_{0_{11,13}} = v_3$$

with the remaining being zero. The v_i are the matrix elements of the Hamiltonian interaction between the atom and the e.m. field in the dipolar approximation, $\delta_i = \omega_i - \omega_{i,i+1} - u_x \omega_1 / c$ is the detuning from resonance and u_x is the velocity of the atom in the direction of the laser beams (supposed to be all in the $\pm x$ direction).

For simplicity we will take $v_1 = v_2 = v_3 = v$. It can be shown, in the three level case, using the results of reference [12] that a departure from this condition strongly distorts the selectivity function $S(\delta_1, \delta_2, \delta_3)$. The distortion is such that to recover high selectivity it is necessary to increase the detuning and this causes a decrease in the ion yield.

The ionization probability of a certain isotope is,

$$\rho_i(\tau, u_x) = \int_0^\tau \gamma_i \rho_{44}(t, u_x) dt \quad (3)$$

where τ is the duration of the ionizing laser pulses (assumed to be synchronized rectangular pulses) and γ_i is the ionization rate.

If we assume a Maxwellian distribution of velocities, the total ion yield is,

$$n_i = \int_{-\infty}^{\infty} du_x f(u_x) \rho_i(\tau, u_x) \quad (4)$$

where $f(u_x) = \frac{1}{\sqrt{\pi}u_T} \exp(-u_x^2/u_T^2)$ and $u_T^2 = kT/m$

The initial condition is $\rho_{11}(t=0) = 1$. The solution for ρ_{44} obtained from equation (1) is,

$$\rho_{44}(t) = i \int_{-\infty}^{\infty} \frac{d\Omega}{2\pi} e^{-i\Omega t} (F^{-1})_{41} \quad (5)$$

where $F(\Omega) = \Omega I - (H_0 + i\Gamma)$, I is a 16x16 identity matrix and the element of the matrix F^{-1} appearing in equation (5) can be written as,

$$(F^{-1})_{41} = \frac{A(\Omega)}{D(\Omega)} \quad (6)$$

In the last equation $A(\Omega) = (Adj F)_{14}$ is the adjoint determinant of the element $(F)_{14}$ and $D(\Omega) = \det F(\Omega)$.

In the present case we are interested in the situation for strong laser fields, thus the conditions

$$v_i \gg \max(\Gamma_{ij}) \quad (7)$$

are satisfied.

Efficient ionization is achieved with long laser pulses, then we assume,

$$\tau \gg 1/v_i \quad (8)$$

The denominator of the integrand in equation (5) has sixteen poles. If equation (7) is satisfied, it is possible to show that under certain conditions to be derived latter (see equations (9)- (15)) twelve of these correspond to highly oscillating frequencies and the four remaining (the relaxation modes) are purely imaginary and of the order of the homogeneous widths Γ_{ij} . Then, taking equation (8) into account, we can neglect the contribution of the highly oscillating modes to $\rho_{44}(t)$. The relaxation mode approximation consists of obtaining the four relaxation modes without computing the oscillating modes.

Let Ω_i , $i = 1, \dots, 16$ be the solutions of the equation $\det F(\Omega) = 0$. We arrange them so that $\Omega_i = -i\lambda_i$, $i = 1, \dots, 4$ (λ_i being real numbers) are the relaxation modes, and Ω_j , $j = 5, \dots, 16$ are the oscillating modes.

Thus, for the relaxation mode approximation to be valid it is necessary that,

$$\Omega_i \gg \max(\lambda_j), \quad i \geq 5, \quad j = 1, \dots, 4 \quad (9)$$

Strictly speaking, equation (9) should be satisfied only for those Ω_i , $i \geq 5$, that populate the upper state. In situations where coherent trapping is possible (like in uranium AVLIS, where a 620 cm^{-1} meta-stable state is involved) some of the highly oscillating modes vanish, but the same occurs with ρ_{44} ($A(\Omega_i) = 0$). Thus, although equation (9) is not satisfied for these Ω_i , the relaxation mode approximation is still valid.

First of all, we will find when equation (9) holds. We analyze the solutions to the equation $D(\Omega) = 0$ in the following cases,

a) $v \gg \delta_j$

It can be shown by direct computation of $D(\Omega)$ that the oscillating modes are,

$$\begin{aligned} \Omega_j^2 &\simeq v^2 & j = 5, \dots, 8 \\ \Omega_j^2 &\simeq 5v^2 & j = 9, \dots, 12 \\ \Omega_j^2 &\simeq (6 + 2\sqrt{5})v^2 & j = 13, 14 \\ \Omega_j^2 &\simeq (6 - 2\sqrt{5})v^2 & j = 15, 16 \end{aligned} \quad (10)$$

b) $\delta_i \gg v_j, \delta_1 + \delta_2 \gg v_j, \delta_2 + \delta_3 \gg v_j$ and $\delta_1 + \delta_2 + \delta_3 \gg v_j$

In this case we have,

$$\begin{aligned} \Omega_j^2 &\simeq \delta_1^2 & j = 5, 6 \\ \Omega_j^2 &\simeq \delta_2^2 & j = 7, 8 \\ \Omega_j^2 &\simeq (\delta_1 + \delta_2)^2 & j = 9, 10 \\ \Omega_j^2 &\simeq \delta_3^2 & j = 11, 12 \\ \Omega_j^2 &\simeq (\delta_2 + \delta_3)^2 & j = 13, 14 \\ \Omega_j^2 &\simeq (\delta_1 + \delta_2 + \delta_3)^2 & j = 15, 16 \end{aligned} \quad (11)$$

c) $\delta_i \gg v, \delta_1 + \delta_2 \gg v, \delta_1 + \delta_2 + \delta_3 \ll v$

Now the oscillating modes are,

$$\begin{aligned} \Omega_j^2 &\simeq \delta_1^2 & j = 5, \dots, 8 \\ \Omega_j^2 &\simeq \delta_2^2 & j = 9, 10 \\ \Omega_j^2 &\simeq (\delta_1 + \delta_2)^2 & j = 11, \dots, 14 \\ \Omega_j^2 &\simeq \frac{\delta_2^2 v^4}{\delta_1^2 (\delta_1 + \delta_2)^2} & j = 15, 16 \end{aligned} \quad (12)$$

d) $\delta_2, \delta_3 \gg v, \delta_1 \ll v, \delta_2 + \delta_3 \ll v$

In this case we can show that,

$$\begin{aligned} \Omega_j^2 &\simeq \delta_2^2 & j = 5, \dots, 10 \\ \Omega_j^2 &\simeq v^2 & j = 11, \dots, 14 \\ \Omega_j^2 &\simeq 4v^2 & j = 15, 16 \end{aligned} \quad (13)$$

e) $\delta_2 \ll v, \delta_1, \delta_3 \gg v, \delta_1 + \delta_3 \ll v$

Now we have,

$$\begin{aligned} \Omega_j^2 &\simeq \delta_1^2 & j = 5, \dots, 12 \\ \Omega_j^2 &\simeq 4v^2 & j = 13, 14 \\ \Omega_j^2 &\simeq \frac{4v^6}{\delta_1^4} & j = 15, 16 \end{aligned} \quad (14)$$

f) $\delta_3 \ll v, \delta_1, \delta_2 \gg v, \delta_1 + \delta_2 \ll v$

In this case we obtain,

$$\begin{aligned} \Omega_j^2 &\simeq \delta_1^2 & j = 5, \dots, 10 \\ \Omega_j^2 &\simeq v^2 & j = 11, \dots, 16 \end{aligned} \quad (15)$$

In cases c) and e) we see that $\Omega_j, j = 15, 16$ decreases with increasing δ_1, δ_2 . For the relaxation mode

approximation to be valid in these two cases, equation (9) restricts the values of δ_1, δ_2 to,

$$\left| \frac{\delta_1 (\delta_1 + \delta_2)}{\delta_2} \right| \ll \frac{v^2}{\max(\lambda_i)} \quad (\text{case } c)$$

$$\delta_1^2 \ll \frac{2v^3}{\max(\lambda_i)} \quad (\text{case } e) \quad (16)$$

Using equation (9) it is possible to simplify the determinants $D(\Omega)$ and $A(\Omega)$ at small frequencies obtaining a fourth and third order polynomials for $D(\Omega)$ and $A(\Omega)$ respectively,

$$\begin{aligned} D(\Omega) &\simeq D_4 \Omega^4 + i D_3 \Omega^3 - D_2 \Omega^2 - i D_1 \Omega + D_0 = \\ &= D_4 (\Omega + i \lambda_1) (\Omega + i \lambda_2) (\Omega + i \lambda_3) (\Omega + i \lambda_4) \\ A(\Omega) &\simeq i A_3 \Omega^3 - A_2 \Omega^2 - i A_1 \Omega + A_0 \end{aligned} \quad (17)$$

We will not give here the expressions for D_i and A_i because they are very large polynomials¹.

From equations (3), (5) and (17) we obtain,

$$\rho_i = \frac{\gamma_i}{D_4} \sum_{j=1}^4 \frac{A(-i\lambda_j)(1 - \exp(-\lambda_j\tau))}{\lambda_j \prod_{k \neq j} (\lambda_k - \lambda_j)} \quad (18)$$

For simplicity, we take the ionization rate to be the only non-vanishing decay rate, thus,

$$\Gamma_{4,4} = 2\Gamma_{j,j} = -\gamma_i, \quad j = 9, 10, 13, 14, 15, 16 \quad (19)$$

the remaining elements of the matrix Γ being zero. We stress the fact that the assumption made for Γ has no effect on the coefficients D_4 and A_3 in equation (17) because they depend only on v_i and δ_j , thus the conclusions we obtain in the next section for the short pulse case are not affected by the form taken by Γ .

From equation (18) it is easy to show that $\rho_i \rightarrow 1$ when $\tau \rightarrow \infty$. This is obtained from the fact that $D_0 = \gamma_i A_0$. In the case under consideration we have,

$$D_0 = \gamma_i^4 v^{12}, \quad A_0 = \gamma_i^3 v^{12} \quad (20)$$

III. The case of short pulses

In the limit of short pulses, when τ is small so that $\exp(-\lambda_i\tau) \simeq 1 - \lambda_i\tau$ (but still large enough to satisfy equation (8)) we have,

$$\rho_i = \gamma_i \tau A_3 / D_4 \quad (21)$$

From the previous equation we can show in the limit of large detuning ($\delta_i \gg v$), that the optimum condition to avoid a decrease in the ion yield of the selected isotope is,

$$\delta_1 + \delta_2 + \delta_3 = 0 \quad (22)$$

To obtain this result we computed the following expression,

$$f(\delta_1, \delta_2, x) = \rho_i(\delta_1, \delta_2, \delta_3) \Big|_{\delta_3 = -\delta_1 - \delta_2} - \rho_i(\delta_1, \delta_2, \delta_3) \Big|_{\delta_3 = -\delta_1 - \delta_2 - \epsilon} \quad (23)$$

In equation (23) we parameterize δ_1 and δ_2 in the following way,

$$\begin{aligned} \delta_1 &= \delta \cos \phi \\ \delta_2 &= \delta \sin \phi \end{aligned} \quad (24)$$

Instead of showing the analytic form of $f(\delta_1, \delta_2, x)$, we rather plot that expression in Fig. 1 for the case $\delta = 40v$, $x = 0.1v$. We see in this figure that f is positive everywhere with minima at $\phi = \pi/4$ and $\phi = 3\pi/4$. Then we conclude that the condition (22) should hold

to avoid a decrease in the ion yield.

Now we investigate the selectivity as a function of δ_1 and δ_2 with δ_3 being fixed by the condition (22).

Let Δ_1, Δ_2 and Δ_3 be the isotopic shifts of the first-step, second-step and third-step atomic transitions respectively for the undesired isotope, then the ion yield for the undesired isotope is,

$$n_i^{(u)} = \rho_i(\delta_1 + \Delta_1, \delta_2 + \Delta_2, -\delta_1 - \delta_2 + \Delta_3) \quad (25)$$

¹ they are available by e-mail

whereas for the selected isotope is,

$$n_i^{(s)} = \rho_i(\delta_1, \delta_2, -\delta_1 - \delta_2) \quad (26)$$

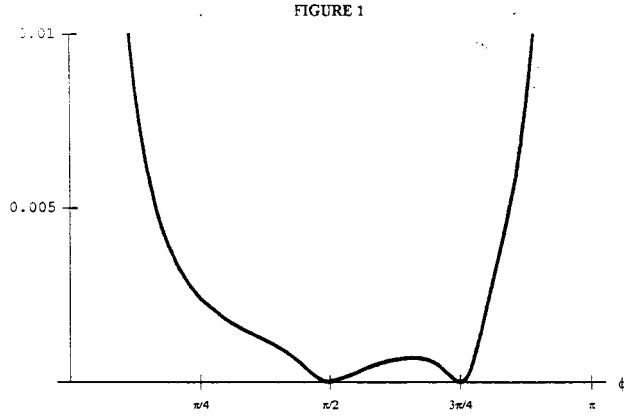


Figure 1. $f(\delta \cos \phi, \delta \sin \phi, x)/(\gamma_i \tau)$ versus ϕ with $\delta = 40v$, $x = 0.1v$ (see equation (23)).

We now consider four cases,

case 1) $\delta_1 \gg v, \delta_2 \gg v, \delta_1 + \delta_2 \gg v$.

In this case we obtain from equations (21), (25) and (26),

$$\begin{aligned} n_i^{(s)} &\simeq \gamma_i \tau \frac{2v^2}{\delta_2^2} \\ n_i^{(u)} &\simeq \gamma_i \tau \frac{2v^6}{\delta_1^2 (\delta_1 + \delta_2)^2 \Delta^2} \end{aligned} \quad (27)$$

where $\Delta = \Delta_1 + \Delta_2 + \Delta_3$

From equations (27) we obtain the selectivity $S = n_i^{(s)}/n_i^{(u)}$,

$$S \simeq \frac{\delta_1^2 (\delta_1 + \delta_2)^2 \Delta^2}{\delta_2^2} \quad (28)$$

which is an increasing function of δ_1 and δ_2 if $\delta_1 \simeq \delta_2$.

case 2) $\delta_1 \ll v, \delta_2 \gg v$

In this case we obtain,

$$\begin{aligned} n_i^{(s)} &\simeq \gamma_i \tau \frac{3v^2}{2\delta_2^2} \\ n_i^{(u)} &\simeq \gamma_i \tau \frac{2v^4(3v^2 + \Delta^2 - \Delta\Delta_1 + \Delta_1^2)}{\delta_2^2(v^2 - \Delta^2 + \Delta\Delta_1)^2(4v^2 + \Delta_1^2)} \end{aligned} \quad (29)$$

Then,

$$S \simeq \frac{3(v^2 - \Delta^2 + \Delta\Delta_1)^2(4v^2 + \Delta_1^2)}{4v^4(3v^2 + \Delta^2 - \Delta\Delta_1 + \Delta_1^2)} \quad (30)$$

which does not depend on δ_2 . If $v \gg \Delta_1, \Delta_2$, then $S \rightarrow 1$, thus there is no selectivity in this case.

case 3) $\delta_2 \ll v, \delta_1 \gg v$

We have in this case,

$$\begin{aligned} n_i^{(s)} &\simeq \gamma_i \tau / 2 \\ n_i^{(u)} &\simeq \gamma_i \tau \frac{2v^6(4v^2 + \Delta^2 + \Delta_2^2)}{\delta_1^4 \Delta^2(4v^2 + \Delta_2^2)} \\ S &\simeq \frac{\delta_1^4 \Delta^2(4v^2 + \Delta_2^2)}{4v^6(4v^2 + \Delta^2 + \Delta_2^2)} \end{aligned} \quad (31)$$

case 4) $\delta_1, \delta_2 \gg v, \delta_3 \ll v$.

In this case $\delta_1 \simeq -\delta_2$. Here we show only the expressions in the limit $v \gg \Delta_j$. The result is,

$$\begin{aligned} n_i^{(s)} &\simeq \gamma_i \tau \frac{3v^2}{2\delta_1^2} \\ n_i^{(u)} &\simeq \gamma_i \tau \frac{3v^2}{2\delta_1^2} \\ S &\simeq 1 \end{aligned} \quad (32)$$

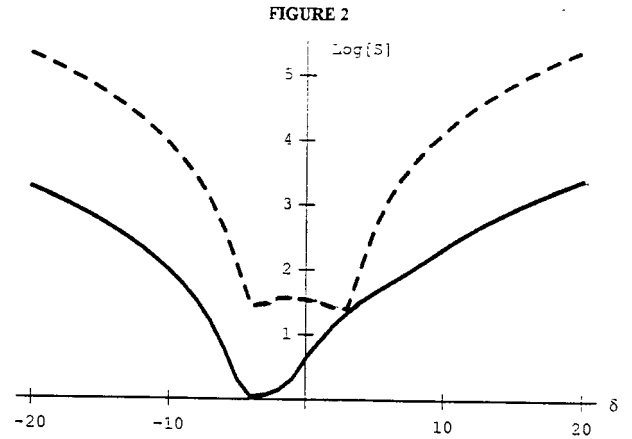


Figure 2. Log plot of the selectivity S in case 3. $\delta_1 = \delta, \delta_2 = 0, \delta_3 = -\delta$. The dashed curve corresponds to the short pulse case (equation (21)). The solid curve corresponds to equation (18). The values of the parameters are $v = 1$ GHz, $\gamma_i = 10^{-3}$ GHz, $\Delta_1 = \Delta_2 = 2$ GHz, $\Delta_3 = 1$ GHz and for the solid curve $\tau = 45$ ns. δ measured in GHz.

We conclude that the most favorable situation is that of case 3 in which selectivity increases as δ_1^4 and the decrease in the ion yield of the selected isotope has a lower bound of $\gamma_i \tau / 2$. This situation changes when the laser pulse duration is increased. In Fig. 2 we show the selectivity for case 3. The dashed curve corresponds to the short pulse approximation whereas the solid curve is for a long pulse (calculated with equation (18)). Fig. 3 shows the selectivity for case 1. We see that for the short pulse case (dashed curves), and for large detuning, the most favorable condition is that of case 3. This situation is reversed for long pulses (solid curves). From the point of view of the ion yield of the selected isotope, the conditions of case 3 are the most favorable in both

the short and long pulse cases. In Fig. 4 we show the ion yield in case 1 (long pulse) where we see a strong decrease for large detuning. This is not so in case 3, where we obtain $n_i \geq 0.99$ in the range $-20 < \delta < 20$.

We note that the two cases where there is no selectivity at large detuning (cases 2 and 4) correspond to the minima at Fig. 1.

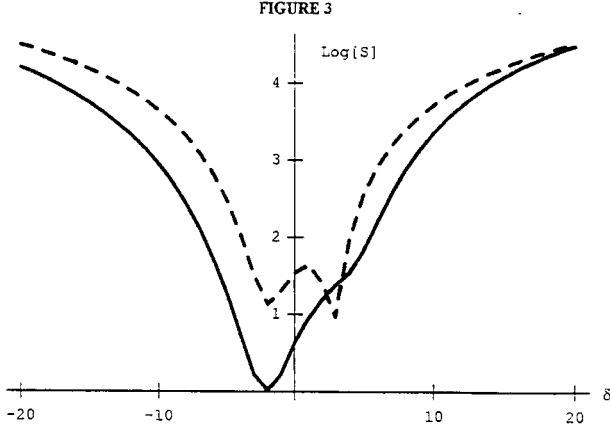


Figure 3. The same as figure 2 for case 1. $\delta_1 = \delta$, $\delta_2 = \delta$, $\delta_3 = -2\delta$.

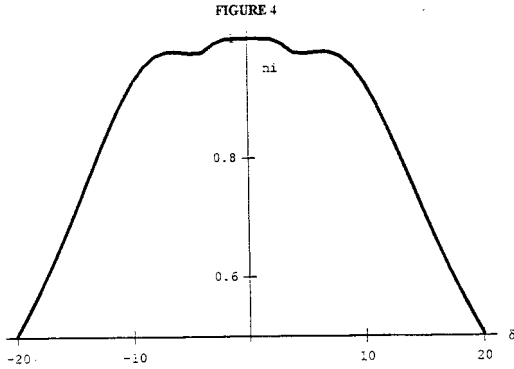


Figure 4. Ion yield of the selected isotope from equation (18). $\delta_1 = \delta$, $\delta_2 = \delta$, $\delta_3 = -2\delta$. $v = 1$ GHz, $\gamma_i = 10^{-3}$ GHz, $\tau = 45$ ns.

IV. Doppler broadening

We will take into account the effect of Doppler broadening when the atoms have a Maxwellian velocity distribution in the case of short pulses and large detuning. We will restrict here to the situation in which $\delta_2 = 0$, $\delta_1 = -\delta_3 = \delta$. As we saw in the previous section, this appears to be the optimum situation (case 3). The remaining cases can be treated in a similar way.

We consider the three laser beams propagating in the $+x$ or $-x$ directions. For an atom with a velocity $\vec{u} = (u_x, u_y, u_z)$ the detunings from resonance are,

$$\begin{aligned}\delta_1 &= \Delta_1 + \delta - k_1 u_x \\ \delta_2 &= \Delta_2 - k_2 u_x \\ \delta_3 &= \Delta_3 - \delta - k_3 u_x\end{aligned}\quad (33)$$

Here $k_i = \pm\omega_i/c$ can be positive or negative depending on the direction of propagation of the laser beams. We take $k_i > 0$ if the corresponding beam propagates in the $+x$ direction and negative otherwise. For the case of the selected isotope we should set $\Delta_1 = \Delta_2 = \Delta_3 = 0$ in equation (33).

First we replace equation (33) in (21) and take the limit of large δ . After this we take the limit $v \gg \Delta_i - k_i u_x$. The resulting expression for ρ_i is,

$$\rho_i = \gamma_i \tau v^6 / P \quad (34)$$

where,

$$\begin{aligned}P &= 2v^6 + c_1 c_2 c_3 v^2 \delta + c_2 c_3 v^2 \delta^2 + c_1 c_3^2 \delta^3 + c_3^2 \delta^4 / 2 \\ c_1 &= (\Delta_1 - \Delta_3) - (h_1 - h_3)z \\ c_2 &= (2\Delta_1 + 3\Delta_2 + 2\Delta_3) - (2h_1 + 3h_2 + 2h_3)z \\ c_3 &= (\Delta_1 + \Delta_2 + \Delta_3) - (h_1 + h_2 + h_3)z\end{aligned}\quad (35)$$

Here we have introduced $z = u_x/u_T$ and $h_i = k_i u_T$.

Replacing (34) in equation (4) we have,

$$n_i = \frac{\gamma_i \tau v^6}{\sqrt{\pi}} \int_{-\infty}^{\infty} dz \frac{e^{-z^2}}{P} \quad (36)$$

The denominator P in the integrand of equation (36) is a polynomial of third degree in z . It can be shown that the two extrema have the following expressions for large detuning,

$$\begin{aligned} z_0 &= \frac{\Delta}{h} + \frac{(h_1 \Delta_2 - h_2 \Delta_1 + h_3 \Delta_2 - h_2 \Delta_3) v^2}{\delta^2 h^2} + O(\delta^{-3}) \\ z_1 &= \frac{\delta}{3(h_1 - h_3)} + O(\delta^0) \end{aligned} \quad (37)$$

where $h = h_1 + h_2 + h_3$.

From (37) we can see that due to the presence of e^{-z^2} we can approximate P in equation (36) by a quadratic expansion near z_0 as a consequence of the fact that $z_1 \rightarrow \infty$ when $\delta \rightarrow \infty$. In the limit $v \gg \Delta_i$ this expansion has the following form,

$$P \simeq 2v^6 + \frac{1}{2} h^2 \delta^4 (z - z_0)^2 \quad (38)$$

Finally, from equations (38) and (36) we obtain,

$$n_i \simeq \frac{\gamma_i \tau}{2\pi} \int_{-\infty}^{\infty} dy; \frac{e^{-(y+z_0)^2}}{1 + \frac{h^2 \delta^4}{4v^6} y^2} \quad (39)$$

Following the analysis made in reference [12] for the three level case, we define the parameter,

$$\kappa = \frac{h\delta^2}{2v^3} \quad (40)$$

The relative ion yield for the selected isotope, obtained from equation (39) is,

$$n_i^{(s)} = \gamma_i \tau \sqrt{\pi} \frac{v^3}{h\delta^2} \quad (41)$$

For the relative ion yield of the undesired isotope we must distinguish three different regimes. First, when $\kappa \ll 1$ we recover the short pulse case; the Doppler broadening is masked by the power broadening and we obtain equations (31) in the limit $v \gg \Delta_i$.

The second and third regimes corresponds to the regime $\kappa \gg 1$. In this case the ion yield for the undesired isotope can be expressed as follows,

$$n_i^{(u)} = \gamma_i \tau \frac{2v^6}{\Delta^2 \delta^4} + \gamma_i \tau \frac{\sqrt{\pi} v^3}{h\delta^2} e^{-\Delta^2/h^2} \quad (42)$$

When $\kappa \gg \frac{h^2}{\Delta^2} e^{\Delta^2/h^2}$, the second term dominates equation (42), then the selectivity approaches,

$$S = e^{\Delta^2/h^2} \quad (43)$$

We see in this situation that the selectivity is independent of the detuning and the ion yield of the selected isotope decreases with δ^{-2} (see equation (41)).

Finally, in the region $1 \ll \kappa \ll \frac{h^2}{\Delta^2} e^{\Delta^2/h^2}$ the first term dominates equation (42) and the selectivity is,

$$S = \frac{\sqrt{\pi} \delta^2 \Delta^2}{2 h v^3} \quad (44)$$

These conclusions have been compared with numerical simulations for long pulses ($\tau = 45$ ns). We will not show figures for these cases. Instead we summarize the results.

We performed simulations for $T = 2000$ °K in the case of uranium. One can show that the selectivity in case 3 (section 3) is greater than in case 1 for large detuning with short pulses. This situation is reversed for long pulses, like in the case without Doppler broadening. However, the ion yield in case 3 decrease much slower than in case 1. We also performed simulations for different directions of the laser beams in case 3. These showed that the most favorable case has one beam propagating in opposite direction to the other two beams. There is no great difference between the three possible combinations. When the three beams propagate in the same direction, the selectivity is slightly smaller than

in the case of one beam reversed. The same is true for the ion yield.

V. Conclusions.

We studied the selectivity and ion yield in a four level system subject to high power resonant laser fields. This study is suitable for application to isotope separation of heavy atoms. We found the conditions to achieve high selectivity without a strong decrease in the ion yield of the selected isotope. We also determined the limits imposed on the selectivity by the Doppler broadening studying the short pulse case. In general, we can say that the condition to avoid a decrease of the ion yield of the selected isotope is $\delta_1 + \delta_2 + \delta_3 = 0$. Constrained to this two dimensional surface in the detuning space, we showed that there are two regions without selectivity: $\delta_1 = 0$ and $\delta_3 = 0$. The most favorable situation in the short pulse case is $\delta_2 = 0$. For longer pulses the situation changes slightly, being $\delta_i \gg v$ more favorable than the case $\delta_2 = 0$ for the selectivity. However, the condition $\delta_2 = 0$ has always the greatest ion yield of the selected isotope. We stress that these results corresponds to the case of at least two large detunings. In the case of small detunings, there are local maxima with greater ion yield. But these points have selectivity much smaller than in the case of large

detunings and seem to be of no interest for industrial applications.

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