Chapter 21

Parameters of Laser Atom Interaction and Photoplasma Relevant for Medical Isotope Separation

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21.1 Introduction

Recently laser-based separation of isotopes, having applications in the field of medicine (Therapeutic and Diagnostic) and industry have attracted a lot of interest, for isotopes such as Yb-176, Yb-168, Lu-177, Sm-153 and other isotopes. Lu-177 and Sm-153 radioisotopes are produced in reactors through (n, γ) reaction followed by β-decay. The medical isotope Lu-177 has two routes of production either from the precursor isotope Yb-176 or from Lu-176. Both these precursors are natural occurring isotopes. Similarly, for the production of Sm-153 radioisotope, the precursor isotope is the naturally occurring low abundant Sm-152 isotope. The purity of the precursor isotopes should preferably be much above 80% for avoiding additional impurity in the generated medical radioisotope and also to have high specific activity for medical applications. More often than not, the enriched precursor isotopes are needed to ensure high specific activity and purity of the radioisotope. Unlike the requirements in nuclear industry that require the production of relatively large quantities (of the order of a tonne per year) of a low enriched (~3-6%) isotope (like U-235), the radioisotope for medical applications must be highly enriched but much lower quantities (few tens of grams) are only needed. Laser-based isotope separation of target isotopes with narrowband lasers are lucrative method of producing high purity precursor isotopes for medical applications.

The precursor isotope Lu-176 is even yet it has non-zero nuclear spin due to odd number of protons and odd number of neutrons, hence hyperfine splitting of each atomic level occurs due to non-zero nuclear spin (I=7). Frequency separation between the most intense hyperfine components of Lu-176 from its neighbouring hyperfine component of Lu-175 should be chosen for selective photoexcitation followed by ionisation puts constraint on the laser spectrum and the process throughput.

In order to generate, efficient laser-based isotope separation with high selectivity, various atomic and laser parameters, requisite interaction and the separation processes of the

targeted ion from the transient photoplasma to the collector plates must be optimized. The existing copper vapor laser pumped wavelength tunable dye laser facility efficiently generates laser radiation in the range 560 nm to 650 nm which correspond to energy about 2 eV. Hence for ionising isotopes with near 6 ev first ionisation energy, three-colour, three-step resonant laser excitations are implemented. Alternatively, two-colour resonant two-step photoionization schemes using the fundamental and second harmonic tuning range of Ti:sapphire laser pumped by high repetition rate DPSS laser are being investigated and reported. Advantage of this system is all solid state laser facility based on Ti:sapphire systems and expected to occupy less lab space. The ionization efficiency and selectivity of the multi-step resonant excitation / ionization scheme are intricately dependent both on the laser parameters like spectral bandwidth, repetition rate, average power, polarization and pulse duration and the atomic parameters like excitation cross-section of the transitions, photoionization cross-section, lifetimes of various levels and their branching ratios, magnetic sub-level degeneracy, isotope shifts, hyperfine structure constants and the atomization source temperature. Fig-

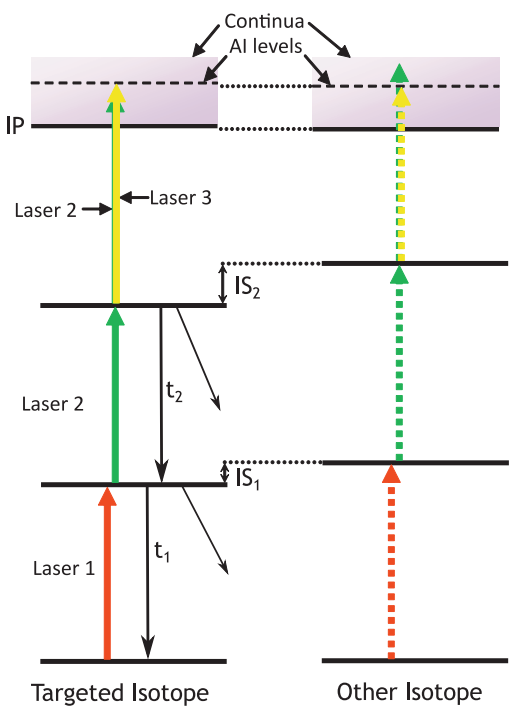

Figure 21.1: Schematic of a three-colour three-step laser excitation and ionisation of the targeted isotope.

ure 21.1 shows the schematic of the three-colour, three-step excitation scheme used in LIS process. Here, t_1, t_2 are the radiative lifetimes along with other branching, $IS_{1,2}$ are level isotope shifts, IP is ionization potential, and AI is autoionization. Branching out of the levels within the excitation ladder will lead to loss of atoms for further excitation as these atoms are lost due to their relaxation into other unconnected metastable states.

21.2 Spectroscopic Parameters for LIS

Three lasers with distinct wavelengths with desired optical powers for three-colour three-step resonant ionisation scheme are required for efficient LIS process utilizing CVL pumped Dye lasers. Third level need to be a suitable autoionization level from higher ionisation cross section view point. Generally power requirement of the first excitation (bound-bound atomic transition) is lower compared third laser as the saturation flux required is more for third-step transition. Second-step transition requires moderate power.

Isotopic shift are due to the change in potential energy seen by the electron due to the mass difference and volume difference of the isotopic nuclei of interest. Generally, for medical isotopes, IS will be around 1-3 GHz. Laser spectral width has to be less than IS for selective excitation of targeted isotope.

Hyperfine components are due to further splitting of fine structure components of an atom due to interaction of effective angular momentum of electron with nuclear spin. Nuclear spin will be zero for even (Z)-even (A-Z) isotopes which will result in single component for each J component. Eg: U-238, Sm-152, and Yb-176. Saturation energy $\frac{h\nu}{\sigma}$ and cross section (σ) of each step is an important parameter for identifying the minimum power intensity required at the laser-atom interaction zone in a process.

Generally, **lifetime** of first excited states will be few hundred nanoseconds and second excited states will have < 100 nsec. Lifetime is an important parameter which decides the pumping rate (σI) and rise time parameter of laser. Here I is peak power intensity of the laser.

There are various line broadening mechanisms that are involved for the spectral width of an atomic line transition, namely natural broadening (1/ lifetime), collisional broadening (related to collisional frequency) and Doppler broadening. In LIS process, Doppler broadening dominates over the other two. Effective Doppler width $\Delta\nu = \Delta\nu_0 Sin\frac{\theta}{2}$, where $\Delta\nu_0$ is the Doppler width depends on the source temperature and atomic mass; θ corresponds to the full angle of divergence of atomic vapour after passing through beam collimator.

$$\Delta\nu_0 = 2\sqrt{ln2}\sqrt{\frac{8k_BT}{mc^2}}\nu_0 = 7.17\times10^{-7}\nu_0\sqrt{\frac{T(K)}{m(amu)}} \tag{21.1}$$

(For Samarium, half angle of 12°, and at $\lambda = 600$ nm, the effective Doppler width is 198 MHz at 800 °C.) Judicious selection of laser power for the first and second steps are required so that power broadening of transition which affects the spectral selectivity of the excitation. Power broadening is the kind of level broadening above Doppler width by an amount related to the ratio of laser intensity to saturation parameter.

Power broadening:

$$\Delta\nu = \Delta\nu_0\left(1+\frac{I}{I_s}\right),$$
$$I_s = \frac{h\nu}{\sigma\tau},$$

where σ is the cross section and τ is the lifetime. Due to power broadening, neighbouring isotopes also gets excited, even if the laser spectral widths are narrower.

21.3 Laser Parameters Recommended from RIS Experiments

Wavelengths for three-step LIS are to be selected from optimal spectroscopic scheme for high ionisation yield with good spectral selectivity. Spectral width of the laser shall be less than that of isotopic shift with target and neighbouring isotopes and HFS components. Laser spectral width also need to be comparable to Doppler width of atomic vapour at interaction zone. Lowest spectral width of the pulsed laser comes from the Fourier limit the pulse width of the laser. For example, 20 nsec dye laser pulse will have 50 MHz for the assumption time bandwidth product constant of 1. For Gaussian spectrum, the constant value is 0.44. Generally single mode laser assumed to have Lorentzian spectral width due to cavity decay time broadening. Single pulse laser spectral width is more important than long time-averaged spectral width as the process interaction is single pulse based. Laser power for saturating transition for first and second excitations should be chosen without affecting selectivity due to power broadening.

Temporal parameters such as pulse width, jitter and rise time are the important parameters for the separation process. Shorter pulse width will have higher peak intensity may help in saturating transition with relatively lower average power or pulse energy. Pulse widths have to be much longer than temporal jitter of laser so that timing among each excitation pulses follows successively from 1st excitation followed by 2nd and then third. Timing delay is also an important factor considering for shorter lifetime of second level, the loss of population may occur for longer delay. Pumping rate (σI) of the levels decides ideal time delay required among the lasers. There are also pulse propagation effects also dictates certain pulse widths for each transition. For example, isotopes having near resonance with target isotopes produces a dispersive component to propagating laser and slow down the pulse. This effect are predominant for first step as it has highest value of diploe moment. This introduces delay among the otherwise synchronised lasers and deteriorate the ionisation yield along the path length. Alternative methods and configurations were discussed in literature to increase the ionisation yield for longer path length. There are other coherent propagation effects, if the laser atom interactions are coherent, which introduces frequency chirping, pulse steepening and soliton formation through self-induced transparency. Sharper rise time always preferred for effective population transfer from one atomic level to higher.

Spatial overlap for longer distance among three laser beams addressing three transitions is an important parameter. Since all three lasers used for LIS process, generated from different MOPA systems have different numbers of amplifier stages and power densities, all will tend to have different beam propagation parameters. Beam size and divergence management ensures to bring all laser beams propagating with good spatial and angular overlaps over a longer interaction length. For good spatially overlapped beam over longer distance, the photon utilization economy will be better for laser atom interaction.

Spectral stability is the key parameter for the high selective excitation. Spectral jitter should be much less than IS and single pulse laser spectral width. Ideally for LIS, it is pulse to pulse interaction dictates LIS process and hence short term spectral width and their spectral jitter will be an important parameter for high yield production with good selectivity.

Power stability may not be an important parameter as all the atomic transitions will be well saturated, especially for first two excitations. Power stability around 5% will be more suitable.

21.4 Laser Atom Interaction – Coherent vs Incoherent

Laser atom interaction, broadly divided into two types of interactions, namely coherent and incoherent. In an incoherent interaction, the population flow monotonically from one level to higher level. For coherent laser interaction, population oscillates at Rabi frequency $\Omega = (\vec{d}.\vec{E})/h$ between the two levels for resonant excitation. Where $\vec{d}$ is the atomic dipole moment and $\vec{E}$ is the electric field of the laser pulse. Incoherent interactions are handled by population rate equation (PRE) which are basically intensity-based interactions and coherent interactions are handled by density matrix equations (Laser field amplitude-based interactions with atomic coherence). Coherent interaction may lead to population trapping among the levels along with other coherent propagation effects. Also using coherent interaction, complete population transfer from one level to next is possible. For a practical system, laser with certain spectral width, coherent interactions decided by the coherence time of laser, interaction time (almost equal to pulse width T_p) and the decay rate of dipole moment of atoms. Interaction time is either laser pule width or transit time of atoms across the laser diameter, whichever is shorter. Decay rate of dipole moment depends on the spectral width of the atomic levels. For atomic dipole relaxation time T_d may be defined as the inverse of atomic spectral width specified in frequency units. If $T_p \ll T_d$, coherent interaction dominates and density matrix to be incorporated. For Medical isotope separation using lasers with single pulse single mode spectral width 60 MHz (laser coherence time = 16 nsec), laser pulse width of 20 nsec and the Doppler width of 250 MHz (T_d ~4 nsec) has time scales in a comparable region. Interactions may not be considered fully incoherent. If one considers lifetime of first (> 100 nsec) and second (50-100 nsec) excited levels, interactions become more coherent for homogenous spectral interactions. Again, interactions depend on the decay rate of autoionization level, if the pumping rate of third excitation is more than of lower transitions, then again interaction converted to incoherent interaction. With the given process parameters, if population pulsation not observed in density matrix formalism, implies the atomic coherence may be ignored and simple rate equations are adequate for process descriptions. Generally, excitation of third level to autoionisation were considered as incoherent, irreversible and handled by rate constant.

21.5 Plasma Parameters

The plasma generated in LIS process is of special category as they are produced by nanosecond laser pulses generating plasma densities varying from $10^6 - 10^{10}$ per cc. Plasma decays to the boundaries and the electrodes with a lifetime varying from few microseconds to 100 microseconds depending on plasma densities and geometries. In LIS process the ions are extracted from the photoplasma by electrostatic methods using parallel plate extraction configuration or their variants. The fields applied for ion extraction were varying from 150 V/cm to 750 V/cm between the plates. As the outermost electron ionised from the target atom, singly ionised target atom is produced along with equal number of ions and electrons. Depending on the plasma density, the charged particles accelerated in the field either follows single particle kinetics or plasma collective kinetics. Electron carries the difference in the energies of three photon and the first ionisation energy of the atom due to lighter mass compared to the target ion. This energy corresponds to electron temperature (T_e). Inside plasma quasi neutrality is maintained corresponds to $n_i = n_e = n$. Two important scaling parameters of plasma are the plasma frequency (Time scaling) and Debye length (length scaling). Plasma frequency (generally for electrons, which will be high due to lighter mass compared to ion) is related plasma density and Debye length depends on both plasma density

and plasma temperature. The relations are given below.

$$\omega_{pe} = \sqrt{\frac{ne^2}{m\varepsilon_0}} \tag{21.2}$$

$$\lambda_D = \sqrt{\frac{\varepsilon_0 k_B T_e}{2ne^2}} \tag{21.3}$$

$$\omega_{pe} = 56.38\sqrt{n\ (m^{-3})} \tag{21.4}$$

$$\lambda_D = 69\sqrt{\frac{T(K)}{n\ (m^{-3})}} \tag{21.5}$$

When Debye length is of the order of one of the dimensions of plasma, the charge collection will show plasma behaviour. Number of particle in Debye sphere shall be much more than 1 qualifies collective behaviour of plasma. Debye length shields the plasma from external electric potential. Table 21.1 lists plasma frequency and Debye length for various plasma densities. Here, T_e = 0.2 eV is assumed. As the photoplasma is generated between biased plates, some percent of electrons will escape the plasma and leave the plasma at higher potential. Further movement of electrons is hindered by net positive potential of the plasma. Near cathode, ion sheath is formed and near anode, electron sheath is formed. Electrons will never reach cathode as it has to climb the potential near cathode. Ion collection time increases from the time of flight ion from plasma edge to higher value as the plasma density increases. Field will not able to penetrate beyond few Debye lengths. Since the plasma has acquired a net positive potential above anode, the plasma will drift slowly towards anode also and deposits ions at anode also for higher plasma densities above $\sim 3\times10^9$ per cc.

Table 21.1: Plasma frequency and Debye length for various plasma densities.

Plasma density	**Plasma angular frequency (electron) rad/sec**	**Debye length (mm)**
1.00E+09	1.78E+09	0.105
5.00E+09	3.99E+09	0.047
1.00E+10	5.60E+09	0.033
5.00E+10	1.26E+10	0.015

Total ion collection time for ion extraction geometry depends on the distance between electrodes, location of plasma, volume of plasma, extraction voltage of the cathode and importantly plasma density. In general, a scaling law for ion extraction time with reference to one particular chosen standard configuration, is observed to follow as given below.

$$\frac{\tau}{\tau_s} = \left(\frac{V}{10^3}\right)^{\alpha} \left(\frac{D}{1}\right)^{\beta} \left(\frac{n_i}{10^{15}}\right)^{\gamma} \tag{21.6}$$

Where V is the extraction voltage, D is the plasma width in cm, n_i is the ion density in m^3. The constants of the exponents can be found by least square fitting of one variable with other two parameters constant. For example, voltage variation for low plasma density corresponds to α of -0.5 and for sheath formation and extraction current based on Child-Langmuir law will have α of -3/2 (for space charge limited flow). These are important factors for ion extraction system behaviour to the sensitive variables.