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High-pressure continuously tunable CO_2 lasers and molecular laser isotope separation

E RONANDER[1,*], H J STRYDOM[1] and L R BOTHA[2]
[1]Klydon (Pty) Ltd, P.O. Box 1552, Pretoria, South Africa
[2]National Laser Centre, P.O. Box 395, Pretoria, South Africa
*Corresponding author. E-mail: einar.ronander@klydon.co.za

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Abstract. The acronym MLIS (molecular laser isotope separation) defines the laser process whereby the isotopes of uranium can be separated by mid-infrared laser/s when the molecule employed is UF_6. The theoretical and spectroscopical data to configure and enable experiments and demonstrations in the laboratory is adequate. However, the engineering and commercial aspects require innovative technology solutions that are not presently available in the literature on these topics. This paper is an overview of the most salient features of MLIS and its potential utility at an industrial level.

Keywords. Molecular laser isotope separation; infrared lasers; UF_6; continuously tunable; isotopic mix.

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1. Introduction

The acronym MLIS (molecular laser isotope separation) has been applied to label the laser process whereby the isotopes of uranium can be separated by mid-infrared laser/s when the molecule employed is UF_6; the latter being the gas on which the current front-end nuclear fuel cycle is based that was established over the past decades requiring a large monetary investment. It, therefore, seems prudent to base any new approaches for the uranium enrichment step on UF_6 unless exceptional commercial reasons may be put forward that merit another advance.

2. Different modes of MLIS

The topic of MLIS of uranium, or any other isotopic species, can be analysed in a more general sagacity as the different labels that have been attached to the versions merely

identify the method by which the desired isotopic species is harvested subsequent to the initial isotopic selectivity. Depictions, e.g. MLIS, CHEMLIS, CRISLA, and SILEX [1] processes all have several common features:

(1) The process gas is UF_6 in combination with a carrier gas.
(2) Laser systems that instigate the isotopic selective step interact with the infrared absorption energy modes of the UF_6 molecule; more specifically the v_3 symmetrical vibrational mode.
(3) The process gas is adiabatically flow-cooled to temperatures below 100 K, in some version well below this temperature, to enhance the spectral purity of the uranium molecule to be able to attain higher laser selectivity.
(4) An initial isotopic infrared selective excitation step at relatively low laser energy fluence to limit the power broadening of the molecular moiety's absorption quality.
(5) Further vibrational excitation by the laser to affect significant dissimilarity in the internal energy content of the desired and undesired isotopic species followed by the different approaches to harvest the product and waste material.

Each of these options, to accomplish the isotope separation of uranium using UF_6, favours one or more technical aspects that may promote the commercial attractiveness of the route. However, in practise the elegant features often become an optical engineer's nightmare. These approaches require temperatures in the laser irradiation zone that can range from 10 to 100 K [2]; and the molecular density that may be realized without noteworthy condensation of the uranium moiety at the operating temperature, covers an equally 'large' range. The original MLIS version where the UF_6 molecule is dissociated to harvest the desired isotope can be accomplished at a uranium density of the order of 10^{15} molecules per cm^3; this implies an irradiation zone length of approximately 5 m for a million separative work units (SWU) per annum plant. At a density of 10^{13} molecules per cm^3 the optical path length, for the same separation capacity, increases by two orders. There is a clear engineering preference for a high working pressure; it would reduce the absorption path length, pumping requirements, and makes the whole concept of mass transfer easier. It is, furthermore, imperative to maintain very good spatial overlap between the laser beam profile and the geometrical distribution of the uranium in the flow-cooled molecular beam to attain effective separation parameters; this constraint can readily be met over single metre distances but is extremely technologically challenging over several hundred metre distances. The infrared beam must be optically engineered and reformed many times over long distances and from experience the only optical elements that can be utilized, in an industrial environment, are molybdenum reflective mirrors that exhibit a minimum reflective loss of 2% per single reflection at best. For example, if 50 reflective optical mirrors are utilized, a number that is low in plant application, 63.6% of the input laser energy will be 'pumped' into heating of the mirrors. Add to this the efficiency of spatial overlap between laser beam and molecular beam, and one can conclude that over 90% of the expensive infrared photons are simply lost in processes other than isotope separation. It is, therefore, essential to strive for a high molecular density in the irradiation zone, even at the expense of other elegant features of the variants for the molecular route of isotope separation.

The desirable laser isotope separation mode should produce high selectivity and enhanced cut for the optimum economic performance. In the first isotope selective irradiation step the laser energy and fluence must be maintained at relatively low values to instigate maximum selectivity; however, this being a resonant vibrational transition the absorption cross-section is high, which in turn induces a fast decrease in the number of laser photons in the beam. This phenomenon is detrimental to the plant design; it is consequently highly preferable to use multiphoton resonant laser excitation in selective step [3].

The v_3 vibration mode of SF_6 and UF_6 has been studied extensively and a substantial amount of data exists on the laser excitation of this molecule in the lower levels. It was demonstrated that apart from the anharmonic shift and the rotational level structure, also of importance is the octahedral splitting of the levels of the triply degenerate $v_3$vibration. To illustrate this, figure 1 presents the structure of the lower vibrational levels of the SF_6 molecule that is a spherical top-type structure that allows for anharmonicity and splitting. In figure 2, the measured two-photon absorption spectrum for SF_6 illustrating the resonances is plotted [4]. The two-photon transitions are much narrower than the single-photon transitions; the latter is power-broadened by the strong laser field whilst for the two-photon transitions this effect is much less. It is highly beneficial for the laser isotope process to utilize the two-photon transitions in the selective laser isotope step.

3. Laser requirements for MLIS

The lasers utilized for laboratory experimentation to study the MLIS mode of isotope separation and that required for industrial scale differ substantially and the latter presents technological challenges far beyond the realms of the laboratory. The method that may be selected to generate the oscillator energy is impacted largely by the ability to operate the

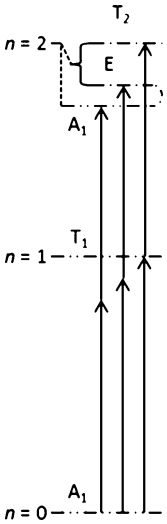

Figure 1. Vibrational energy splitting for SF_6.

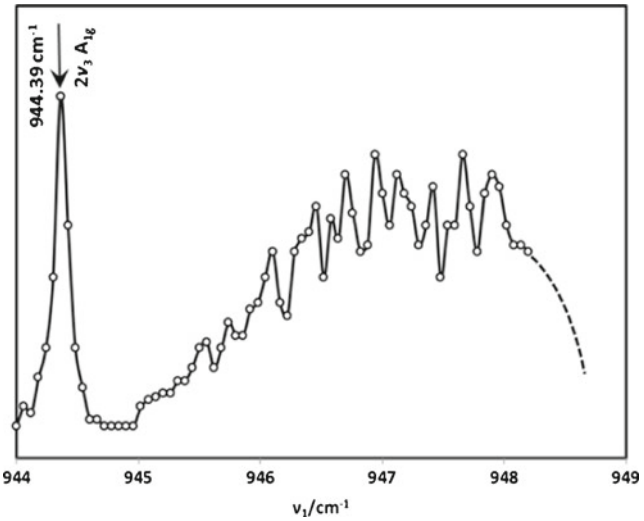

Figure 2. Measured two-photon transitions for SF_6 [4].

laser at pulse repetition frequencies of 1,000 to 2,000 Hz, as single units, at high levels of output stability, laser subsystems and components performing to the extended lifetimes that are imperative in an isotope separation plant.

The strong symmetrical stretch vibration of the UF_6 molecule is the best-suited energy mode for selective isotopic excitation of ^{235}U isotope, and the resonant frequency is in the region of 628 wave numbers. It is practical to select the technologically well-developed mid-infrared laser operating on the CO_2 molecule but first this frequency must be converted to the $\sim$628 cm^{-1} region by utilizing stimulated Raman scattering (SRS) in a para-hydrogen medium [5]. Both the quantum conversion and the energy conversion efficiencies of this translation are very good and the process is scalable to high repetition frequencies. A second non-linear frequency conversion technique can be based on four-wave mixing between the Nd:YAG laser and the CO_2 laser; whilst the SRS is performed at liquid nitrogen temperature, the four-wave mixing translation operates well at ambient temperature.

4. High-pressure continuously tunable CO_2 laser

Continuous tunability of a CO_2 laser can be realized by utilizing the pressure broadening of the discrete transitions in the CO_2 laser lines. When a laser is operated on a single isotopic gas composition, e.g. $C^{16}O_2$, the pressure required for continuous wavelength output in the R28 to R32 of the 10.6 μm band transitions, demands absolute pressures greater than 10 atm [6]. Such tunability has been reported in the scientific literature but these systems present a number of technical challenges that limit the scalability to industrial-type lasers. Figure 3 plots the wavelength tunability of a very highly-pressure CO_2 laser to illustrate the laser gain at these elevated conditions.

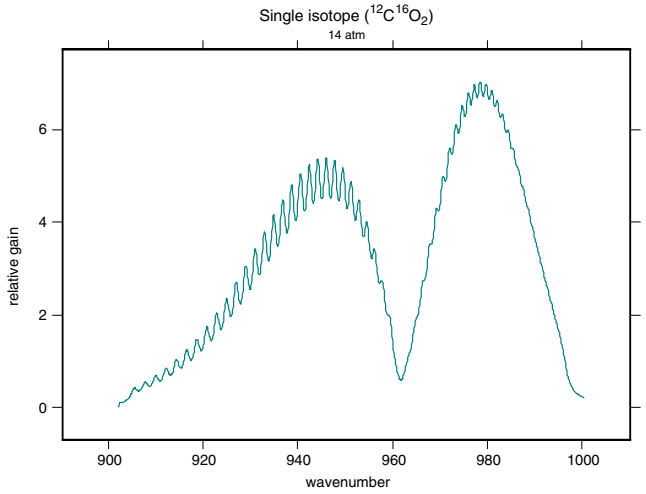

Figure 3. Wavelength tunability for CO_2 laser at high pressure.

The obvious inadequacies of the high-pressure laser can be collated:

(1) The design and manufacture of a pressure vessel demands special codes and is costly.
(2) It is tricky to sustain a stable electrical discharge and control acoustic shock waves in the laser discharge cavity at high pulse repetition frequencies.
(3) Very high electrical voltages are needed for the discharge to be triggered; typically 10 kV/cm electrode gap per atmosphere pressure.
(4) Transport of the laser gas mixture in the laser vessel for high repetition frequencies.
(5) The peak intensity of laser energy inside the laser cavity reaches extremely elevated levels during lasing build-up and the lifetime of the partial reflector in the laser cavity will be accordingly short.

Figure 3 illustrates the same type of laser gain plot for a mixture of 25% $C^{16}O_2$, 50% $C^{16}O^{18}O$, and 25% $C^{18}O_2$; a more smooth gain curve at a lower pressure is possible with the isotopic mix [7]. Figure 4 summarizes the laser output energy for a constructed 2.7 atm total pressure isotopic mix laser with the output energy adequate for the selective step in MLIS.

5. High repetition frequency lasers

The optical engineer is faced with several technological challenges to scale the output lasing performance of the single laboratory units, and to assemble a laser chain that can operate continuously under industrial demand for many months where the lifetimes of components are measured in many billions of laser shots. The cost balance point between capital investment and operational cost converges to single units functioning at ∼2000 Hz pulse repetition frequency. At these pulse rates, a billion laser shots accumulate in ∼140 h.

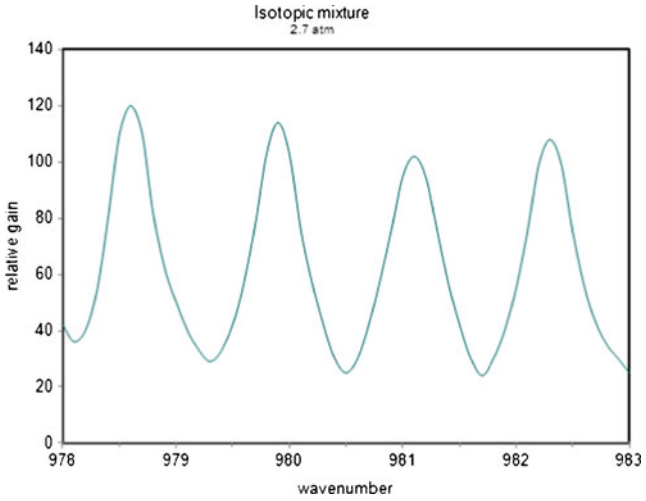

Figure 4. Wavelength tunability of isotopic mix CO_2 laser at 2.7 atm.

A summary of the focal domains that support lasers that operate at these high work rates is: the ultrafast electronic switching of the input energy into a gas glow discharge; the prolonged lifetimes of these switching components; prevention of even the slightest onset of arcing between the laser electrodes; the attainment of sufficient high-quality aerodynamic flow over laser electrodes; ultralow time jitter of the optical pulse from the laser cavity; extended lifetimes of the optical elements in the laser resonator and chain configuration.

The pulse amplitudes and durations necessary to be achieved for the laser switching, lie in the range where the switch speeds and other parameters of semiconductors are inadequate and where the working life of conventional gas discharge apparatus is drastically reduced by the extreme switching demands. Pulse compression is an elegant technique to transfer the burden of excessively high current densities and unmanageable current rise rates from semiconductor switches to electromagnetic switches. Current compression in time is accomplished through series resonance in capacitors and saturable inductors in a transmission line configuration. Electrical energy is transferred in this process from one capacitor stage to the next, with a reduction in transfer time in each successive stage and a commensurate increase in current amplitude. These pulses can be as short as 50 to 100 ns, and may have amplitudes in the kiloampere and kilovolt ranges [8].

In addition to the design and construction of suitable pulse compression subsystems, also crucially important is the development of voltage-controlled pulse power supply suitable to charge the initial storage energy capacitors, and which sensitively determines the eventual time jitter of the optical laser pulses. Figure 5 showcases the type of power supply developed for this purpose as well as the first industrial-type compressor for a 2000 Hz CO_2 laser [8]. The lifetimes of these subsystems can be quoted in terms of years rather than months. Currently, the market status is that mode-switched power supplies that operate at very high (40 kHz) electrical pulses that charge the storage energy unit, can be procured on the open supply market, and combining these with pulse compression the time jitter on the optical pulses is less than 1 ns. The latter is crucial for efficient energy output and pulse stability of the Raman cell.

Figure 5. Power supply units and four-stage current pulse compressor unit for high laser pulse frequency operation.

Figure 6 shows a 2000 Hz CO_2 laser chain comprising all three wavelengths in the UF_6 irradiation scheme cited here in a demonstration unit to confirm continuous operation and lifetime testing. The Raman conversion step was based on a cold cell, at liquid nitrogen temperature, with design that deviated from the well-known Herriot cell (figure 7); the

Figure 6. Three-wavelength laser chain operating at 2000 Hz for MLIS application.

Figure 7. Raman cell for stimulated Raman scattering conversion at 2000 Hz.

optical beams never cross as in the better known version as this can be a source of sporadic behaviour and laser amplitude fluctuation. The λ_2 wavelength is the only one generated from SRS, whilst the lower energy λ_1 is generated from a four-wave-mixing interaction.

The phenomenon of electrical arcing between the laser electrodes can be the laser engineer's biggest nightmare and may be labelled as the Achilles Heel of gas discharge lasers. Electrical arcing in gas discharge lasers must be defined more comprehensively than

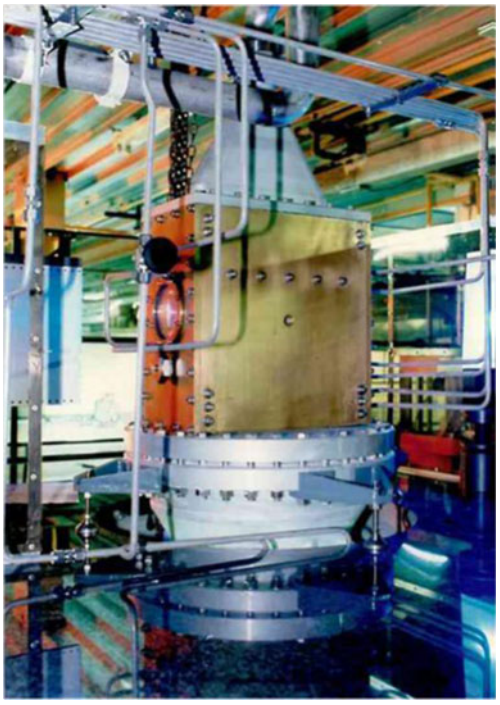

Figure 8. Long path length flow cooling nozzle configuration.

simply the 'short circuit' concept commonly associated with arcing; even small pressure variations between the electrodes or local weak acoustic waves give rise to current variations that soon develop into preferential current channels that destroy electrode surface at the very high laser pulse repetition frequencies. The solution is found in innovative segmented electrode profiles' efficient pre-ionization techniques, shock wave suppression, and inventive gas flow patterns between the laser electrodes.

6. Laser/gas interaction

It is crucially important to finagle an optimal overlap between optical laser beam and the spatial distribution of gas molecules in the irradiation zone; this aspect can distinctly influence the economics for the better or worse. When a large polyatomic molecule, e.g. uranium hexafluoride is used, the need for substantially cooling the quantized energy modes of the molecule is imperative for isotope separation. The application of free jet expansion to establish low temperatures is of limited practical use as the geometrical dimension cannot be straightforwardly controlled which beckons a decreased overlap between the two distributions.

We found the Laval-type nozzle expansion much more efficient to flow cool large molecules, and the flow channel with cooled gas can be largely controlled in dimensions [9]. It is, furthermore, relatively trivial to force equilibrium amongst the vibrational, rotational, and translational degrees of freedom of the target molecule. In figure 8, a 1 m Laval nozzle for UF_6 flow cool and irradiation is showcased. An overlap of better than 80% in volume distribution between laser photons and cooled molecules was obtained in this design. The vibrational temperature that may be attained in this nozzle type is sufficient to attain single-step enrichment factors to reactor-grade fuel.

7. Summary

This paper overviews some of the important features of the laser isotope separation of uranium in the MLIS mode. Information on this topic is controlled by the International Atomic Energy Agency (IAEA) protocols and country specific legislation. The cited references contain accessible data and information on the highlighted aspects of importance.

The essential science that underpins the separation of isotopes by laser is adequately demonstrated and recorded. However, the translation to industrial and commercial exploitation presents challenges that have not yet been described in literature. This paper discusses some of the 'obvious' solutions that have a major impact on the technology and the industrial viability of MLIS as an isotope separation process.

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