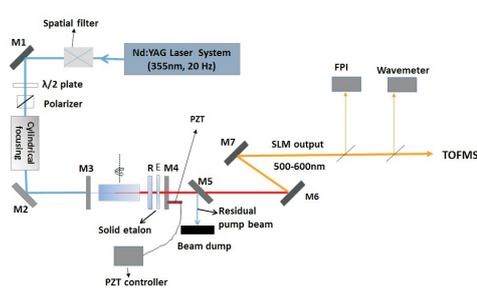


Isotope Enrichment

Development and Application of Single Longitudinal Mode Optical Parametric Oscillator for Selective Photoionization Studies on Lu-176 Isotope

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Schematic of the SLM OPO experimental setup

ABSTRACT

We have designed and developed a single longitudinal mode (SLM) OPO (Optical Parametric Oscillator) pumped by the third harmonic of Nd:YAG laser operating at 20Hz repetition rate. The SLM OPO is continuously tunable over a spectral range of 500–600nm and exhibits a spectral linewidth ($\Delta\nu$) of ~ 200 MHz over this entire spectral range. The developed SLM OPO has been integrated with the TOFMS (Time-of-flight Mass Spectrometer) setup and used as an alternative first step excitation source to the existing three-step selective photoionization scheme used for selective photoionization studies on Lu-176 with multi-mode dye lasers. The photoionization studies reveal that just by using the SLM OPO in place of multi-mode dye laser ($\Delta\nu \sim 2$ GHz) as first step excitation source, the degree of enrichment of Lu-176 is enhanced from $\sim 85\%$ to better than 96% .

KEYWORDS: Single Longitudinal Mode (SLM), Optical Parametric Oscillator (OPO), Selective Resonant Photoionization, Enrichment of Lu-176.

Introduction

Optical Parametric Oscillators (OPOs) are highly efficient devices for generating tunable coherent radiation in the visible to IR range[1]. They can cover the frequencies which are not directly accessible with existing conventional tunable lasers. The attractive features of these devices include high power, all-solid-state compact design, and the possibility of a very broad wavelength tuning range making them an ideal choice for use in basic and applied research as well as industrial, environmental, and healthcare applications, where laser tunability is essential.

Laser-based selective multi-step photoionization and subsequent collection of the desired isotope is a very lucrative separation technology, particularly for medical isotopes where the typical product requirement is in the range of a few milligrams to a gram. To attain high purity in the product, tunable lasers with narrow linewidths (preferably SLM) are essential, particularly in the case of isotopic systems exhibiting overlapping spectra. Further, tunable SLM lasers are desirable for selectivity studies of such isotopes and precise spectroscopic characterization of their atomic levels and transitions involved in the selective photoionization scheme. However, the commercially available SLM tunable lasers, suitable for high-resolution spectroscopy, are too expensive. Further, the conventional narrowband liquid dye lasers widely used for these applications are limited by their wavelength tunability. The tuning range of these dye lasers is typically 25-30nm for a given dye and thus requiring multiple dyes to cover the broad spectral range in the visible region, which is cumbersome and time-consuming. For example, using a

β -BaB₂O₄ (BBO) crystal pumped by the third harmonic of an Nd:YAG laser, the output of the singly resonant OPO is continuously tunable from 412nm to 650nm[2]. However, the inherent shortcoming of the OPO is that the nonlinear-optical phase-matching conditions naturally cause relatively broad spectral linewidth.

Recently, we have developed a new design of narrowband OPO and demonstrated narrowband operation down to SLM by employing cylindrical focusing to the pump and intra-cavity pump beam reflector along with an etalon[3]. Here we present our work wherein this cavity design was improved and incorporated with minor modifications to develop an automated SLM OPO module with wavelength and cavity length controls for wavelength tuning and frequency stabilization. The developed SLM OPO module was combined with the existing laser set up and used in proposed three-step selective photoionization ladder for the enrichment of Lu-176 from its natural composition (Lu-175: 97.41% and Lu-176: 2.59%). The three-step photoionization schemes[4,5] can provide high selectivity and degree of enrichment of $\sim 85\%$ with multimode dye lasers ($\Delta\nu \sim 2$ GHz) at high intensities requisite for high ionization. As a next step, the SLM OPO output was used in place of the multimode dye laser tuned to the first step excitation wavelength and studied the performance by evaluating the enhancement in selectivity and degree of enrichment of Lu-176 isotope.

The SLM OPO Design

The optical configuration of the SLM OPO is shown schematically in Fig.1(a) while Fig.1(b) shows a pictorial view of the experimental setup. The SLM OPO consists of a type-II phase matched BBO crystal with dimensions $6 \times 12 \times 15\text{mm}^3$ (cut: $\theta = 37^\circ$, $\phi = 30^\circ$). The crystal is placed in a plane-plane

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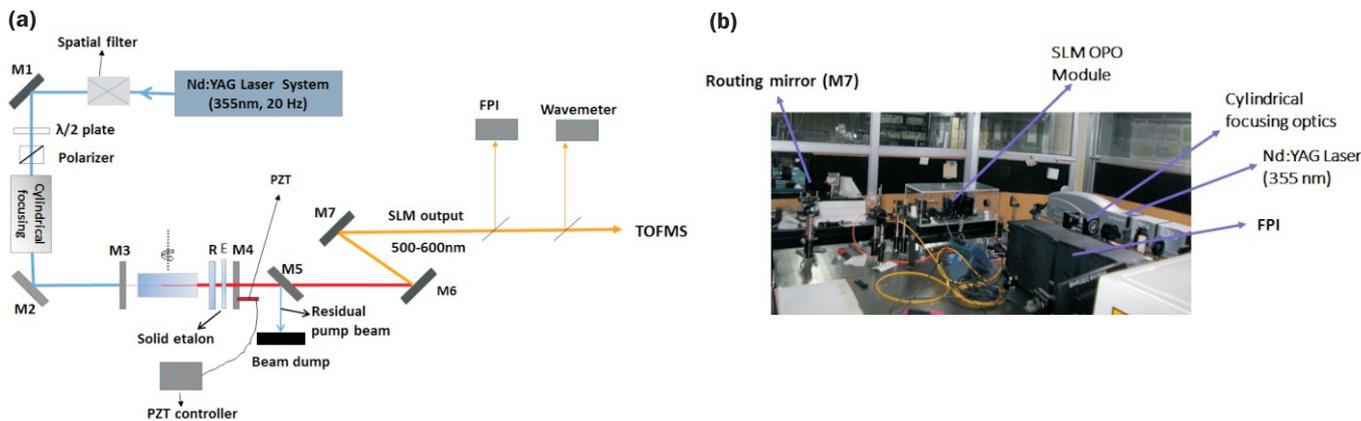


Fig.1: (a) Schematic of the SLM OPO experimental setup
(b) Pictorial view of the experimental setup in the RIMS laser facility.

cavity formed by broadband mirrors labeled as M3 and M4 with reflectivity 99% and 70%, respectively, in the spectral region 490nm to 630nm while at the pump wavelength of 355nm both the mirrors have high transmission (>95%). The BBO crystal was pumped by third harmonic of an Nd:YAG laser (355nm) which delivers a maximum output power of ~400mW with smooth temporal profile at repetition rate of 20Hz and FWHM of ~8ns. A spatial filtering technique has been employed for the pump beam, which removes the spatial intensity variations in the pump beam and generates a near Gaussian intensity profile and a variable attenuator comprising of a half-wave plate and a polarizer to control the pump laser pulse energy. In addition, a cylindrical focusing system[6, 7] has been employed for the pump beam to generate an elliptical beam at the center of the type-II BBO. The cylindrical focusing increases the interaction length of the signal with the pump in the nonlinear crystal and reduces the threshold pump pulse energy required for the OPO.

An intra-cavity pump beam reflector, R, whose reflectivity is ~99% at 355nm (high transmission for signal and idler wavelengths) was placed after the BBO crystal, to double pass the pump beam through the BBO crystal. In type-II phase matched OPOs, the spectral linewidth of the resonant signal wave is predominantly due to the divergence bandwidth of the signal wave (divergence bandwidth much greater than the inherent gain bandwidth) because of the asymmetric wavelength change with the noncollinear angle in Type-II phase matching. However, the double-pass configuration of the pump beam helps in reducing the divergence broadening in which, only the collinear rays of the signal propagating in the direction of the pump wave experience gain at the same wavelengths for both forward and backward pass through the nonlinear crystal. The noncollinear rays experience the gain at different wavelengths in the forward and backward passes because of the asymmetric wavelength change with noncollinear angle, leading to a decrease in effective gain[8]. The double passing has facilitated spectral narrowing[3] by reducing the divergence broadening. As a result, the spectral linewidth has been reduced from 500GHz to ~ 90GHz and also has lowered the oscillation threshold from 2mJ/pulse to 1mJ/pulse. To reduce the spectral linewidth further and to achieve SLM operation of the OPO, a solid Fabry Perot (FP) etalon whose FSR is 79GHz, Finesse 22 was chosen to place inside the OPO cavity after the pump beam reflector, shown as 'E' in Fig.1(a). Furthermore, to provide a better longitudinal mode discrimination for stable SLM operation of the OPO, the length of the cavity has been restricted to well within 4.5cm, corresponding to an optical length of 6cm. Both the crystal and

etalon were mounted on computer-controlled stepper motorized stages with an angular resolution of 1μrad. The coarse wavelength tuning of the SLM OPO is achieved through the rotation of the crystal. The angular rotation of the etalon provides the fine adjustment to the etalon transmission peak for matching with the peak of the OPO cavity gain bandwidth. The output coupler (M4) was mounted on a PZT driven translation for active stabilization of the cavity. The residual pump beam was dumped by using a dichroic mirror M5. The routing mirrors M6 and M7 having high reflectivity at the OPO signal wavelengths were used to route the OPO output to the TOFMS. The SLM OPO output was sampled as shown in the Fig.1(a) for wavelength measurement and monitoring the fringe pattern to ensure the single mode operation of the OPO during the experiment. The wavelength was monitored using a high resolution wavemeter having absolute wavelength accuracy of 30MHz. A Fabry Perot etalon (FSR 7.5GHz) based Interferometer (FPI) was constructed to monitor the fringe pattern of the SLM OPO continuously during experiment. The SLM OPO output was delivered up to the TOFMS chamber of the RIMS experimental set up for selective photoionization experiments on natural lutetium.

Performance of the SLM OPO

The developed Type-II BBO based OPO is continuously tunable from 490nm to 630nm. Fig.2 shows the wavelength

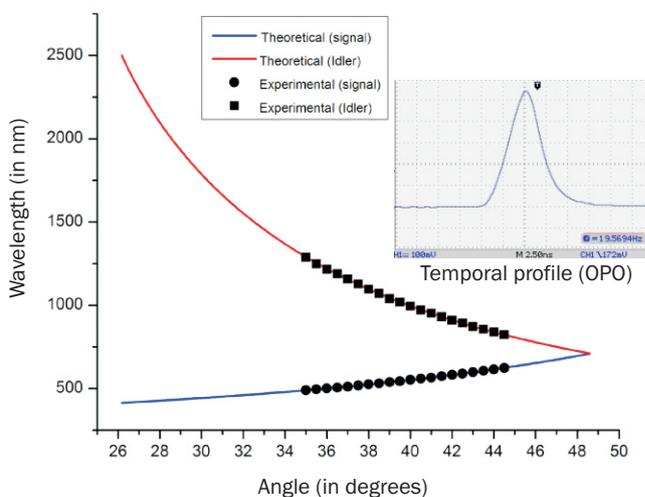


Fig.2: Wavelength tuning curve of the signal wave and the corresponding idler wave with change in internal angle to the optic axis of Type-II BBO OPO. Inset: The oscilloscope trace of the temporal profile of the OPO signal output.

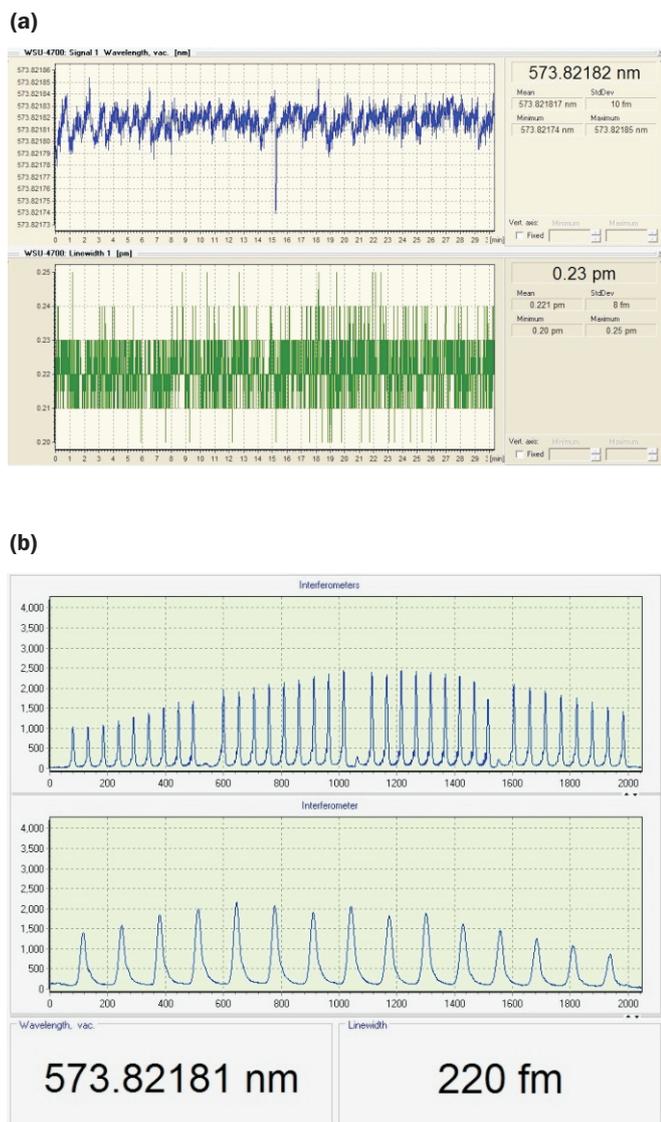


Fig.3: Spectral characteristics of SLM OPO (a) Frequency stability (b) Interferometer fringe pattern of the high resolution Wavemeter.

tuning curve of the signal wave and corresponding idler with a change in internal angle to the optic axis of Type-II BBO crystal. However, the insertion of FP solid etalon in the OPO cavity, which is coated for R~88% in the spectral region of 500-600nm, restricted the tuning range to 500-600nm. The temporal pulse width (FWHM) of the SLM OPO was measured to be ~4.5ns. The oscilloscope trace of the SLM OPO signal output is shown in the inset of Fig.2. High spectral purity of ~200MHz and high wavelength stability of the source laser are requisite for the first step excitation for achieving high selectivity and a high degree of enrichment of the Lu-176. Fig.3(a) shows the long term frequency as well as spectral linewidth stability of the SLM OPO at 573.82181nm measured over a duration of 30 min. The spectral linewidth of the SLM OPO operating at 573.82181nm was measured to be ~220fm which corresponds to ~200MHz. The standard deviation of the wavelength and spectral linewidth during the long term operation was measured to be $\pm 10\text{fm}$ and $\pm 8\text{fm}$ respectively. Fig.3(b) shows the high-resolution wavemeter interferometer fringe pattern of the SLM OPO output. The SLM OPO was set to operate at a power of ~1.5mW at 20Hz repetition rate, sufficient for beam sampling for diagnostics and TOFM.

RIMS Experimental Facility

The RIMS experimental set up for multi-step multi-colour resonance ionization mass spectroscopy[4] comprises of three multi-mode dye lasers DL-1 to DL-3 pumped by second harmonic of Nd:YAG lasers and an indigenously developed Time Of Flight Mass Spectrometer (TOFMS) with mass resolution of ~200amu. The dye laser beams are combined to form a collinear beam with good spatial overlap. Time delay of few nanoseconds can be introduced between the pulses from (DL-1, DL-2) and (DL-2, DL-3), by use of optical delay line or digital delay generator to ensure their sequential arrival at the laser-atom interaction zone of the TOFMS. An atomic beam generator integrated with TOFMS delivers well collimated beam of Lu atoms in the interaction region to spatially overlap with all the three dye laser beams. The photo-ions generated are directed by use of DC electric fields to an MCP detector through the mass analyzer module of TOFMS. Mass spectra thus obtained can be recorded using a digital storage oscilloscope and/or a gated integrator based data acquisition and laser control system. Parts of the lasers are sent to a wavelength meter for wavelength determination.

Selectivity Experiments on Natural Lutetium

We have improved the three-step photoionization scheme for Lu-176 enrichment reported earlier[4] by selecting a stronger albeit broader autoionization resonance at 577nm[5] which reduces the power requirement on ionizing laser. It is depicted schematically in Fig.4, wherein solid arrows indicate the hyperfine pathway chosen for selective photoionization of Lu-176 by virtue of the high selectivity as well as maximum accessibility it offers. Consequently, the selectivity achieved by this scheme with multi-mode lasers DL-1 to DL-3 of line width ~2 GHz tuned to first, second and third step transitions is ~85% at laser powers adequate for enrichment runs.

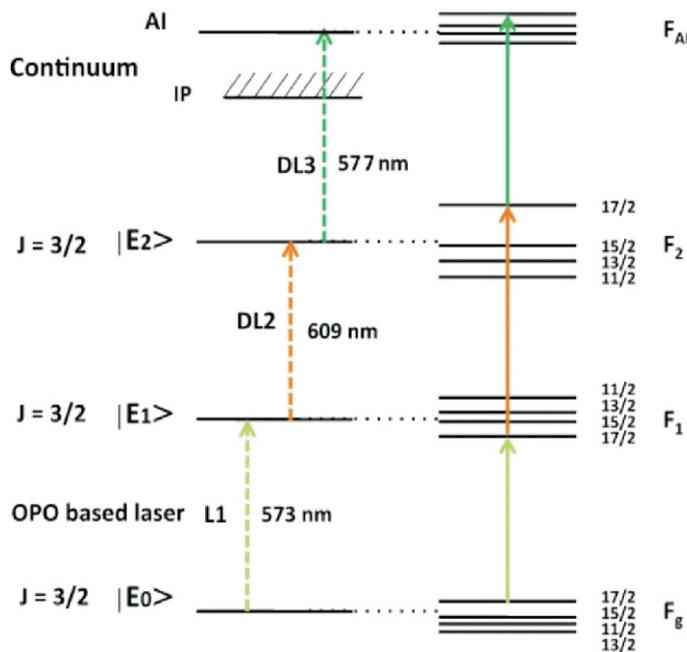


Fig.4: Schematic of three-step three-colour resonance photoionization scheme used for selective ionization of Lu-176. The dotted arrows show the three-step scheme while solid arrows indicate the hyperfine pathway used offering high selectivity and high accessibility.

Effect of SLM OPO on the Isotopic Selectivity of Lu-176

To evaluate the effect of narrow line width on selectivity, the multi-mode laser beam from DL-1 providing first step excitation was replaced by the SLM OPO output beam of comparable power. The SLM OPO was precisely tuned to the $F_g = 17/2$ $F_1 = 17/2$ hyperfine component of the first step transition of Lu-176 at 573nm exciting its population in lower hyperfine level to upper hyperfine level with $F = 17/2$. The multimode lasers DL-2 and DL-3 were tuned as before to the second and third step transition wavelengths of Lu-176 at 609nm and 577nm respectively so as to follow the ionization pathway $F_g = 17/2$ $F_1 = 17/2$ $F_2 = (17/2)$ F_{Ai} . The replacement of multi-mode laser beam from DL-1 by SLM OPO laser beam resulted in remarkable enhancement in the selectivity.

Fig.5 shows mass spectra of Lu isotopes recorded using TOFMS where multi-mode laser from DL-1 (spectrum in black colour) and SLM OPO (spectrum in blue colour) was used for first step excitation. The powers of both DL-1 and OPO were adjusted to 0.1mW measured over cross-sectional area of diameter 3mm. Corresponding powers of DL-2 and DL-3 were 0.3mW and 1.5mW, respectively. These values are comparable to powers intended for use in enrichment experiments. With first step excitation by multi-mode laser, degree of enrichment of ~85% was achieved for Lu-176 from its natural abundance of 2.6%. This corresponds to a separation factor of ~213. With SLM OPO beam of identical intensity, the degree of enrichment increased to >96 %, thus,

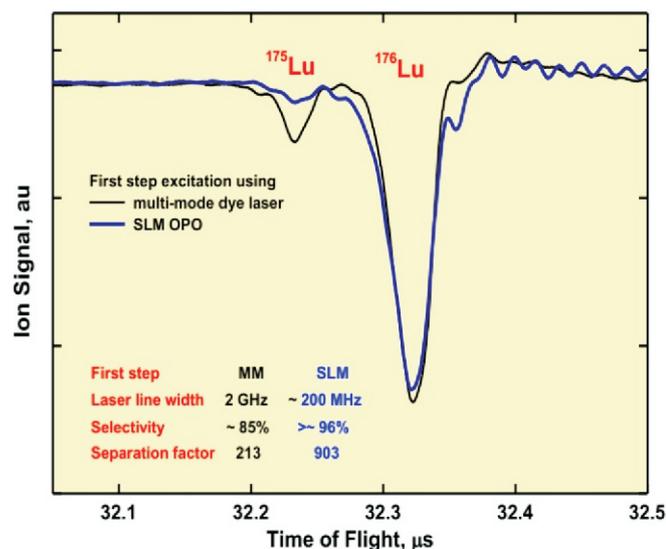


Fig.5: Mass spectrum of Lu isotopes recorded in TOFMS with L1, L2 and L3 tuned to the photoionization pathway $F_g = 17/2$ $F_1 = 17/2$ $F_2 (17/2)$ F_{Ai} corresponding to 573nm, 609nm and 577nm transitions, respectively of ^{176}Lu . The black and blue mass spectra are recorded with first step excitation from multi-mode dye laser ($\Delta\nu \sim 2$ GHz) and SLM OPO laser ($\Delta\nu \sim 200$ MHz), respectively while L2 and L3 are multi-mode ($\Delta\nu \sim 2$ GHz). Comparison of the two spectra shows clear enhancement in selectivity of ^{176}Lu to ~96% when SLM OPO laser is used.

increasing the separation factor to >903. It may be noted that in enrichment experiments, along with first step, when second excitation transition is also provided by SLM lasers, the spectroscopic selectivity at these laser powers is expected to enhance to >99%.

Conclusion and Future Scope

In conclusion, we have developed a single longitudinal mode OPO, which is tunable in the spectral region of 500-600nm. The SLM OPO has been set up in the RIMS laser facility for utilization as a first step excitation source alternative to the existing multimode laser for the selective photoionization studies of Lu-176. The spectral characteristics of the SLM OPO have been studied, and it is found that the time averaged spectral linewidth of the OPO is less than 200MHz in the spectral region of 500-600nm. Employing the SLM OPO as the first step excitation source resulted in substantial enhancement (from 85% to 96%) in the degree of enrichment of Lu-176. Furthermore, considering the fact that the second step selectivity of our photoionization scheme is higher than the first step, these results signify >99% spectroscopic selectivity when single-mode lasers are employed for both the first and second step selective excitation of Lu-176 in the enrichment experiments.

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